



# Microwave-assisted chemical looping gasification of plastics for H<sub>2</sub>-rich gas production

Wenming Fu, Yaning Zhang<sup>\*</sup>, Weitao Cao, Wenke Zhao, Bingxi Li

School of Energy Science and Engineering, Harbin Institute of Technology (HIT), Harbin 150001, China

## ARTICLE INFO

### Keywords:

Microwave-assisted gasification  
Chemical looping gasification  
Plastic  
Gas production  
NiFe<sub>20</sub>O<sub>x</sub> oxygen carrier

## ABSTRACT

Microwave-assisted heating coupled with chemical looping gasification was first proposed for the recycling of plastics into H<sub>2</sub> rich gas, achieving a hydrogen conversion efficiency of up to 84.3%. In the experiment, plastic was initially decomposed into hydrocarbon volatiles, followed by the gasification reactions in the presence of an oxygen carrier under vacuum conditions for 10 min. Subsequently, air was introduced for wax removal and oxygen carrier recovery for another 10 min. This study evaluated the chemical looping gasification performances of polypropylene using oxygen carriers Ni<sub>a</sub>Fe<sub>b</sub>O<sub>x</sub> with different Ni/Fe molar ratios (0, 1:40, 1:20, 1:10, 3:20, and 1:5). NiFe<sub>20</sub>O<sub>x</sub> showed the highest gas yield of 81.3 mmol/g<sub>pp</sub> with an H<sub>2</sub> yield of 46.0 mmol/g<sub>pp</sub>. Among different microwave powers (590 W, 690 W, 790 W, 890 W, and 990 W), 890 W was identified as the ideal microwave power for maximum gas yield and H<sub>2</sub> yield. Microwave-assisted heating exhibited high efficiency, achieving a heating rate of 450 °C/min at 890 W, with the microwave oven consuming only 0.182 kWh of electrical power during the entire experiment. In the chemical looping gasification of various plastics (polypropylene, high-density polyethylene, low-density polyethylene, polystyrene, and plastics mixture) over NiFe<sub>20</sub>O<sub>x</sub> under 890 W microwave power, polypropylene yielded the highest gas and H<sub>2</sub> production. This study provides a novel approach for efficiently recycling plastics, alleviating environmental pollution and contributing to energy sustainability.

## 1. Introduction

Plastic products are ubiquitous in modern society due to their affordability, malleability, and convenience, which have made them exceedingly popular among consumers. As global demand continues to rise, plastic production reached a substantial 400.3 million tonnes annually in 2022 [1]. However, their non-biodegradability and prohibitive recycling costs lead to significant environmental pollution and pose threats to human health [2]. Plastics often end up in landfills or the natural environment, where they can persist for centuries, releasing harmful chemicals and microplastics into ecosystems [3]. The most widely used types of plastics include polypropylene (PP), low-density polyethylene (LDPE), high-density polyethylene (HDPE), and polystyrene (PS), collectively constituting approximately 18.9%, 14.1%, 12.2%, and 5.2% of global plastic production, respectively [1]. This study focuses on these four types of plastics.

Gasification emerges as a promising method for recycling plastic waste by producing combustible gas [4]. However, the generated gas typically has a low heating value due to the inclusion of gasification

agents such as air and carbon dioxide [5]. To address this issue, the chemical looping gasification method is proposed, wherein a solid oxygen carrier replaces conventional gasification agents [6]. The oxygen carrier can be reduced to provide oxygen atoms for the reaction with the plastic vapor, generating gas ( $C_xH_y + MO_z \rightarrow (2x-z) CO + (z-x) CO_2 + (y/2) H_2 + M$ ) [7]. Subsequently, the reduced oxygen carrier can be regenerated by reacting with air or steam [8]. Metal oxides are commonly employed as oxygen carriers due to their cost-effectiveness, high reactivity in redox reactions, and substantial oxygen storage capacity [9]. For instance, Yao et al. [7] reported a carbon conversion efficiency of 99.03% and a gas production of 177.89 mmol/g<sub>pp</sub> with a lower heating value of 11.35 MJ/Nm<sup>3</sup> from the chemical looping gasification of PP.

Chemical looping gasification of solid wastes typically operates at temperatures above 800 °C in an electric heating furnace to achieve high carbon conversion efficiency [10]. However, maintaining such high temperatures consumes considerable energy as electric heating not only heats the feedstocks but also the surrounding air. Microwave-assisted heating offers higher thermal efficiency compared to electrical heating due to its different heating mechanism [11]. Microwave heating utilizes

<sup>\*</sup> Corresponding author.

E-mail address: [ynzhang@hit.edu.cn](mailto:ynzhang@hit.edu.cn) (Y. Zhang).

<https://doi.org/10.1016/j.cej.2024.156225>

Nomenclature			
<b>Abbreviations</b>			
AS	Air introduction stage	$H_p$	is hydrogen content in per gram plastic
EN	Sum of vacuum stage and air introduction stage	$m_0$	is weight of condensers, thermocouple, and connecting tubes before experiment, g
$Fe_3O_4$	Ferrosoferric Oxide	$m_1$	is weight of condensers, thermocouple, and connecting tubes after vacuum stage, g
HDPE	High-density polyethylene	$m_2$	is weight of quartz reactor, SiC, oxygen carrier, and plastic before experiment, g
HHV	Higher heating value	$m_3$	is weight of quartz reactor, SiC, oxygen carrier, and wax after vacuum stage, g
LDPE	Low-density polyethylene	$M_a$	is relative molecular mass of gas component $a$
MP	Mixed plastics	$m_f$	is weight of feed plastic, g
PP	Polypropylene	$v_a$	is the volumetric concentration of gas component $a$ in the vacuum stage, vol. %
PS	Polystyrene	$v_b$	is the volumetric concentration of gas component $b$ in the air introduction stage, vol. %
SA	Collected oxygen carriers after air introduction stage	$v_{N_2}$	is volumetric concentration of nitrogen in the air introduction stage, vol. %
SiC	Silicon carbide	$Y_a$	is specific molar yield of gas component $a$ , mmol/g <sub>plastic</sub>
SV	Collected oxygen carriers after vacuum stage	$Y_b$	is specific molar yield of gas component $b$ , mmol/g <sub>plastic</sub>
VS	Vacuum stage	$Y_{gam}$	is specific molar yield of gas obtained in the air introduction stage, mmol/g <sub>plastic</sub>
<b>Variables</b>		$Y_{gv}$	is gas yield in the vacuum stage, wt. %
$a$	is gas component obtained in the vacuum stage	$Y_{gvm}$	is specific molar yield of gas obtained in the vacuum stage, mmol/g <sub>plastic</sub>
$AN_2$	is the volumetric concentration of nitrogen in the air, vol. %	$Y_{ov}$	is oil yield in the vacuum stage, wt. %
$b$	is gas component obtained in the air introduction stage	$\eta_C$	is carbon conversion efficiency, %
$C_a$	is carbon content in gas component $a$	$\eta_H$	is hydrogen conversion efficiency
$C_b$	is carbon content in gas component $b$		
$C_p$	is carbon content in per gram plastic		
$H_a$	is hydrogen content in gas component $a$		
$H_b$	is hydrogen content in gas component $b$		
$HHV_a$	is the HHV of gas component $a$ , MJ/Nm <sup>3</sup>		
$HHV_g$	is the HHV of gas, MJ/Nm <sup>3</sup>		

electromagnetic waves to generate heat through mechanisms such as dipole rotation, ionic conduction, and interfacial polarization [12]. Moreover, materials exhibit varying microwave absorption capabilities [13], related to factors like dielectric constant, loss tangent, polarity, conductivity, and moisture content [14]. This method directly and uniformly heats materials, minimizing energy losses and enhancing overall efficiency [15]. Furthermore, microwave heating has a higher heating rate, which makes the reactions more vigorous, leading to a faster release of volatiles [16], thereby benefiting gas production [17]. An et al. [18] explored microwave-assisted chemical looping gasification of water hyacinth for syngas production, achieving a cold gas efficiency of 78.66% at 1000 W microwave power. However, research on microwave-assisted heating in chemical looping gasification of plastics remains absent.

Oxygen carriers played a pivotal role in the chemical looping gasification process [7], and extensive research has focused on identifying suitable oxygen carriers [19]. Ideal oxygen carriers should be cost-effective, mechanically stable, and exhibit excellent performance characteristics, including high oxygen transport capacity, high selectivity [20], and superior redox properties [21]. Metal oxides, including Ni, Fe, Mn, Cu [22], W, V [20] and Co [23], have been investigated extensively for chemical looping gasification. Iron-based oxygen carriers, such as  $Fe_3O_4$ , offer several advantages, such as abundant lattice oxygen, multiple oxidation states, and low cost [24]. For example, Xu et al. [25] investigated the chemical looping gasification of rice husk using  $Fe_3O_4$  as the oxygen carrier, achieving a syngas yield of 46.99 mmol/g biomass. Iron oxides also exhibit favorable microwave absorption properties [26], making them compatible with microwave-assisted heating in chemical looping gasification. Fan et al. [13] studied the microwave heating performance of  $Fe_3O_4$ , achieving an instantaneous heating rate of up to 289.2 °C/min at 650 W microwave power, with an average heating rate of approximately 125 °C/min up to 1000 °C. However, Fe-based oxygen carriers are susceptible to sintering, which can negatively impact the gasification process and gas production [22].

To mitigate these challenges, Fe-based oxygen carriers are often modified with other oxides [27]. Additionally, combining two active metals can synergistically enhance catalytic performance beyond that of individual metals [28]. For instance, Qiu et al. [29] demonstrated that  $CoFeO_x$  as an oxygen carrier achieved a  $H_2$  yield of 11.17 mmol/g in chemical looping  $H_2$  production from water splitting, surpassing  $Fe_2O_3$  (7.61 mmol/g). The synergistic effects of Co doping into  $Fe_2O_3$ , validated through temperature-programmed reduction analysis, were shown to promote the reduction of Fe. Huang et al. [30] examined the chemical looping gasification of biomass char using  $NiFe_2O_4$ , which exhibited superior char gasification performance with a carbon conversion efficiency of 88.12%, significantly higher than that of individual  $Fe_2O_3$  or a mechanical mixture of  $Fe_2O_3$  and  $NiO$ , due to the Fe/Ni synergistic effect. Despite these advancements, research on identifying oxygen carriers suitable for microwave-assisted heating in chemical looping gasification of plastics is lacking.

$H_2$  is a clean energy carrier with significant potential to mitigate environmental challenges, and it is the primary gas product of chemical looping gasification of plastics [7]. Key parameters that influence  $H_2$  yield include reaction temperature, microwave power, oxygen carrier type, and plastic type. Reaction temperature plays a crucial role in the chemical looping gasification process, as higher temperatures promote the cracking of large molecules [8]. Previous studies have demonstrated that increasing temperature generally results in higher gas yields and greater  $H_2$  production [31]. Similarly, higher microwave power leads to faster heating, which accelerates the decomposition of plastic waste and promotes the release of hydrogen and other volatiles [32]. However, an optimal balance must be maintained to avoid issues such as localized overheating or inefficient energy use. The selection of a suitable oxygen carrier is critical for facilitating the redox reactions in the gasification process [33]. Modifications to oxygen carriers, such as doping with Ni or Co, can further enhance  $H_2$  production by improving catalytic performance [20]. Additionally, different types of plastics have varying hydrogen content and chemical structures with differing bond energies,

which influence the gasification process and  $H_2$  yield [34]. To the best of the authors' knowledge, no study has investigated the influences of microwave power, oxygen carrier, and plastic type on the microwave-assisted chemical looping gasification of plastics. Therefore, it is of great interest to explore how these factors affect  $H_2$ -rich gas production in this process.

In this study, Ni was incorporated onto  $Fe_3O_4$  through wet impregnation to produce  $Ni_aFe_bO_x$  (where  $a$  and  $b$  represent the Ni/Fe molar ratio). The microwave-assisted heating coupled with chemical looping gasification of plastics was investigated using  $Ni_aFe_bO_x$  as the oxygen carrier to produce high-quality gas. Various Ni/Fe molar ratios (0, 1:40, 1:20, 1:10, 3:20, and 1:5) and different microwave powers (590 W, 690 W, 790 W, 890 W, and 990 W) were examined to optimize the Ni/Fe molar ratio and microwave power, and to explore their effects on gas production. Using  $NiFe_{20}O_x$  with a 1:20 Ni/Fe molar ratio (the optimal oxygen carrier) at a microwave power of 890 W (the optimal microwave power), the microwave-assisted chemical looping gasification of PP, PS, LDPE, HDPE, and mixed plastics (MP) was performed to evaluate their performance in gas production.

## 2. Materials and methods

### 2.1. Materials

The plastics used in this study, including pure PP, LDPE, HDPE, and PS particles, were sourced from China Shenhua Coal to Liquid and Chemical Co., Ltd. Silicon carbide (SiC) obtained from Qinghe Andi Metal Materials Co., Ltd., was used as microwave absorbent with a particle size range of 0.25–0.85 mm. Ferrosoferric Oxide ( $Fe_3O_4$ ), acquired from Nangong Xindun Alloy Welding Material Spraying Co., Ltd., served as the support material for the oxygen carriers due to its widespread availability and excellent microwave absorption properties. Nickel nitrate hexahydrate ( $Ni \cdot (NO_3)_2 \cdot 6H_2O$ ), provided by Sinopharm Chemical Reagent Co., Ltd., was utilized as the nickel source.

### 2.2. Synthesis method of oxygen carries

Oxygen carriers with different Ni/Fe molar ratios of  $Ni_aFe_bO_x$  were synthesized using the wet impregnation method. The preparation procedure involved dissolving a predetermined quantity of  $Ni \cdot (NO_3)_2 \cdot 6H_2O$  in 80 ml of deionized water, followed by the addition of 15.47 g  $Fe_3O_4$ . The mixture was agitated at 80 °C and 1000 rpm for 4 h in a magnetic stirring water bath to ensure homogenization. Subsequently, it was oven-dried at 105 °C for 24 h to evaporate all the water. The dried material was then crushed into a powder and calcined in a muffle furnace under an oxidative atmosphere at 550 °C for 5 h. Then the oxygen carriers were sieved using standard screens, with particle sizes ranging from 0.074 to 0.096 mm.

### 2.3. Experimental procedures

Fig. 1 illustrates the schematic diagram of the microwave-assisted chemical looping gasification system, which comprises a microwave oven, a power meter socket, a gas supply system, a temperature measuring system, a quartz flask reactor, a gas bag, condensers, connecting tubes, and valves. The cavity of the microwave oven (0 ~ 1000 W, 2450 MHz) measures 300 mm in length, 300 mm in width, and 350 mm in height. A 30 mm diameter hole is fabricated on the upper surface of the microwave oven for inserting a connecting tube. A 40 mm high pipe seal is placed above the hole to minimize microwave leakage. The power meter socket was used to measure the microwave oven's electrical energy consumption during the experiment. The gas supply system, which includes a flow meter, an air compressor, and a quartz straight tube, was used to supply air to the quartz flask reactor. The temperature measuring system, consisting of a K-type thermocouple and a digital thermometer, was employed to measure the temperature of the feedstocks. The quartz flask reactor, with a chamber volume of 100 ml, was used to contain the feedstocks. At the quartz flask reactor's outlet, three condensing tubes immersed in an ice-water mixture, were placed in series to cool the heavy volatiles and collect the condensable oil. A gas bag connected to the outlet of the condensers was used to collect the non-condensable gas. Two valves were used to control the reactor environment by opening and closing as needed.

Each experiment was repeated at least twice to ensure data reproducibility. Throughout the study, 1-g plastic, 6-g oxygen carrier, and 12-g SiC were fixed based on the pre-experiments. SiC possessed excellent microwave absorption ability and was used to absorb and convert more microwaves into heat to improve thermal efficiency. The experiment was divided into two stages: the vacuum stage and the air introduction stage. In the vacuum stage, the condensers, thermocouple, quartz straight tube, quartz flask reactor, and connecting tubes were initially weighed. The plastic particles, oxygen carriers, and SiC were weighed separately, then mixed uniformly and placed in the quartz flask reactor. The experimental system was connected as shown in Fig. 1, with insulation cotton filled in the microwave oven to reduce heat loss. Valve 1 was turned off and valve 2 was turned on. The experimental system was vacuumed with a vacuum pump for ten minutes. Then valve 2 was turned off and the vacuum pump was replaced with a gas bag. The microwave oven was started at the designed microwave power. When the temperature of the feedstock reached 900 °C, it was maintained within the range of 880–920 °C by turning the microwave oven on and off. After ten minutes, the microwave oven and valve 2 were turned off, and the gas bag was removed. Once the temperature of feedstock cooled below 100 °C, the condenser tubes, thermocouple, quartz straight tube, quartz flask reactor, and connecting tubes were weighed again. In the air introduction stage, the experimental system was reconnected as shown in Fig. 1. Valve 1 was turned off and valve 2 was turned on. The experimental system was vacuumed for ten minutes, then valve 2 was turned off and the vacuum pump was replaced with a new gas bag. The microwave oven was started again. Once the temperature reached 900 °C, the air compressor, valve 1, and the flow meter set at 50 ml/min were turned on. The temperature was maintained within the range of 880–920 °C by turning the microwave oven on and off. After ten minutes of air inlet, the microwave oven, valve 1, and valve 2 were turned off, and the gas bag was removed. When the temperature cooled below 100 °C, the condenser tubes, thermocouple, quartz straight tube, quartz flask reactor, and connecting tubes were weighed again. Finally, the spent oxygen carrier was collected for characterization, and the gas components in the two gas bags were analyzed using mass spectrometry.

### 2.4. Characterization methods

The produced oil weight was determined by assessing the weight variance of the condensers, thermocouple, and connecting tubes before and after the experiment. The produced gas weight was calculated by

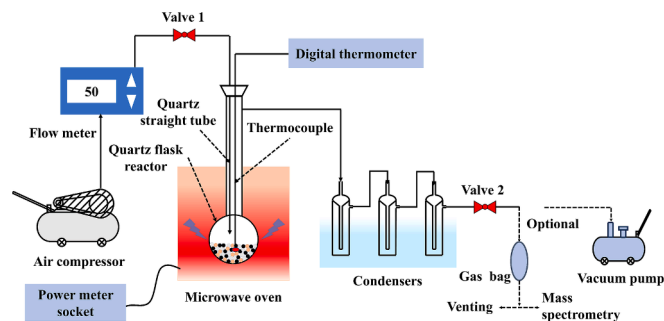


Fig. 1. Schematic diagram of the microwave-assisted chemical looping gasification system.

subtracting the oil weight from the sum of the plastic particles' weight and the weight reduction of the oxygen carriers. These calculations are detailed in Equations (1), and (2).

$$Y_{ov} = \frac{m_1 - m_0}{m_f} \times 100\text{wt.}\% \quad (1)$$

$$Y_{gv} = \frac{m_2 - m_3}{m_f} \times 100\text{wt.}\% - Y_{ov} \quad (2)$$

where:

$Y_{ov}$	is oil yield in the vacuum stage, wt.%
$Y_{gv}$	is gas yield in the vacuum stage, wt.%
$m_0$	is weight of condensers, thermocouple, and connecting tubes before experiment, g
$m_1$	is weight of condensers, thermocouple, and connecting tubes after vacuum stage, g
$m_2$	is weight of quartz reactor, SiC, oxygen carrier, and plastic before experiment, g
$m_3$	is weight of quartz reactor, SiC, oxygen carrier, and wax after vacuum stage, g
$m_f$	is weight of feed plastic, g

The molar gas yield in the vacuum stage and the air introduction stage was calculated using Equation (3) and Equation (4), respectively.

$$Y_{gvm} = \frac{1000Y_{gv}}{\sum_a v_a M_a} \quad (3)$$

$$Y_{gam} = \frac{500A_{N_2}}{22.4m_f v_{N_2}} \quad (4)$$

where:

$a$	is gas component obtained in the vacuum stage
$M_a$	is relative molecular mass of gas component $a$
$Y_{gvm}$	is specific molar yield of gas obtained in the vacuum stage, mmol/g <sub>plastic</sub>
$Y_{gam}$	is specific molar yield of gas obtained in the air introduction stage, mmol/g <sub>plastic</sub>
$A_{N_2}$	is the volumetric concentration of nitrogen in the air, vol.%
$v_a$	is the volumetric concentration of gas component $a$ in the vacuum stage, vol. %
$v_{N_2}$	is volumetric concentration of nitrogen in the air introduction stage, vol. %

The molar gas yield of each gas component in the vacuum stage and air introduction stage was calculated using Equation (5), and Equation (6), respectively.

$$Y_a = Y_{gvm} \times v_a \quad (5)$$

$$Y_b = Y_{gam} \times v_b \quad (6)$$

where:

$b$	is gas component obtained in the air introduction stage
$Y_a$	is specific molar yield of gas component $a$ , mmol/g <sub>plastic</sub>
$v_b$	is the volumetric concentration of gas component $b$ in the air introduction stage, vol. %
$Y_b$	is specific molar yield of gas component $b$ , mmol/g <sub>plastic</sub>

The higher heating value (HHV) of the gas was calculated using Equation (7). The carbon conversion efficiency and hydrogen conversion efficiency were calculated using Equations (8), and (9), respectively.

$$HHV_g = \sum_a HHV_a v_a \quad (7)$$

$$\eta_c = \frac{\sum_a Y_a C_a + \sum_b Y_b C_b}{C_p} \times 100\% \quad (8)$$

$$\eta_H = \frac{\sum_a Y_a H_a + \sum_b Y_b H_b}{H_p} \times 100\% \quad (9)$$

where:

$HHV_g$	is the HHV of gas, MJ/Nm <sup>3</sup>
$HHV_a$	is the HHV of gas component $a$ , MJ/Nm <sup>3</sup>
$\eta_c$	is carbon conversion efficiency, %
$C_a$	is carbon content in gas component $a$
$C_b$	is carbon content in gas component $b$
$C_p$	is carbon content in per gram plastic
$\eta_H$	is hydrogen conversion efficiency
$H_a$	is hydrogen content in gas component $a$
$H_b$	is hydrogen content in gas component $b$
$H_p$	is hydrogen content in per gram plastic

The surface morphology of the raw oxygen carriers was investigated using a Scanning Electron Microscope (SEM). The crystalline composition of the oxygen carriers—including the raw oxygen carriers, the spent oxygen carriers after the vacuum stage, and the spent oxygen carriers after the air introduction stage—was determined using X-Ray Powder Diffraction (Empyrean Intelligent X-ray Diffraction System) over the 2θ range of 0° to 90°, with a step size of 0.167°. It was used to confirm the loading of Ni on Fe<sub>3</sub>O<sub>4</sub> and identify changes in the crystalline components following the vacuum and air introduction stages.

### 3. Results and discussion

#### 3.1. Characterization of oxygen carriers

##### 3.1.1. Surface morphology

The surface morphologies of the oxygen carriers are presented in Fig. 2. SEM images showed that Ni loading did not alter the structural framework of Fe<sub>3</sub>O<sub>4</sub>. Additionally, it was observed that Ni was primarily loaded on the surface of the oxygen carrier, and as the Ni ratio increased, more Ni was encapsulated on the surface of Fe<sub>3</sub>O<sub>4</sub>.

##### 3.1.2. Crystal structure

Fig. 3 shows the XRD spectra of the oxygen carriers. For fresh Fe<sub>3</sub>O<sub>4</sub>, peaks corresponding to Fe<sub>3</sub>O<sub>4</sub> (COD #9009768) were detected. For the collected Fe<sub>3</sub>O<sub>4</sub> after the vacuum stage, peaks corresponding to Fe (#4113941), Fe<sub>3.78</sub>O<sub>4</sub> (#1541150), and Fe<sub>3</sub>O<sub>4</sub> (#9009770) were detected. The appearance of Fe and Fe<sub>3.78</sub>O<sub>4</sub> was attributed to the reactions between Fe<sub>3</sub>O<sub>4</sub> and the generated hydrocarbon vapors, where some lattice oxygens from Fe<sub>3</sub>O<sub>4</sub> combined with the hydrocarbons, producing Fe and Fe<sub>3.78</sub>O<sub>4</sub>. For the collected Fe<sub>3</sub>O<sub>4</sub> after the air introduction stage, peaks corresponding to Fe (#1100108), Fe<sub>3.78</sub>O<sub>4</sub> (#1011168), Fe<sub>2</sub>O<sub>3</sub> (#2108028), and Fe<sub>3</sub>O<sub>4</sub> (#9009768) were detected. The appearance of Fe<sub>2</sub>O<sub>3</sub> was attributed to the oxidation of Fe (#1100108) or Fe<sub>3.78</sub>O<sub>4</sub> (#1011168) by O<sub>2</sub>. For fresh NiFe<sub>20</sub>O<sub>x</sub>, peaks corresponding to NiFe<sub>2</sub>O<sub>4</sub> (#5910064), NiO<sub>2</sub> (#9012319), Fe<sub>2</sub>O<sub>3</sub> (#9015065), and Fe<sub>3</sub>O<sub>4</sub> (#1532800) were detected. The presence of Fe<sub>2</sub>O<sub>3</sub> was due to the oxidation of Fe<sub>3</sub>O<sub>4</sub> by O<sub>2</sub> during the calcination process in the muffle furnace. For the collected NiFe<sub>20</sub>O<sub>x</sub> after the vacuum stage, peaks corresponding to Fe (#9013472), Ni (#9013029), Fe<sub>3.66</sub>O<sub>4</sub> (#1541151), FeO (#9008636), Ni<sub>0.58</sub>Fe<sub>1.42</sub> (#1524199), and Fe<sub>3</sub>O<sub>4</sub> (#9002026) were detected. The presence of Fe, Fe<sub>3.66</sub>O<sub>4</sub>, and FeO was attributed to the reduction of Fe<sub>2</sub>O<sub>3</sub> or Fe<sub>3</sub>O<sub>4</sub> by the generated hydrocarbon vapors, and Ni and Ni<sub>0.58</sub>Fe<sub>1.42</sub> were derived from NiO<sub>2</sub> and NiFe<sub>2</sub>O<sub>4</sub>. For the collected NiFe<sub>20</sub>O<sub>x</sub> after the air introduction stage, peaks corresponding to Fe (#4113941), Ni (#2100652), Fe<sub>2</sub>O<sub>3</sub> (#9000139), FeO (#9008636), and Fe<sub>3</sub>O<sub>4</sub> (#9002321) were detected. The presence of Fe<sub>2</sub>O<sub>3</sub> was due to the oxidation of FeO or Fe by O<sub>2</sub>.



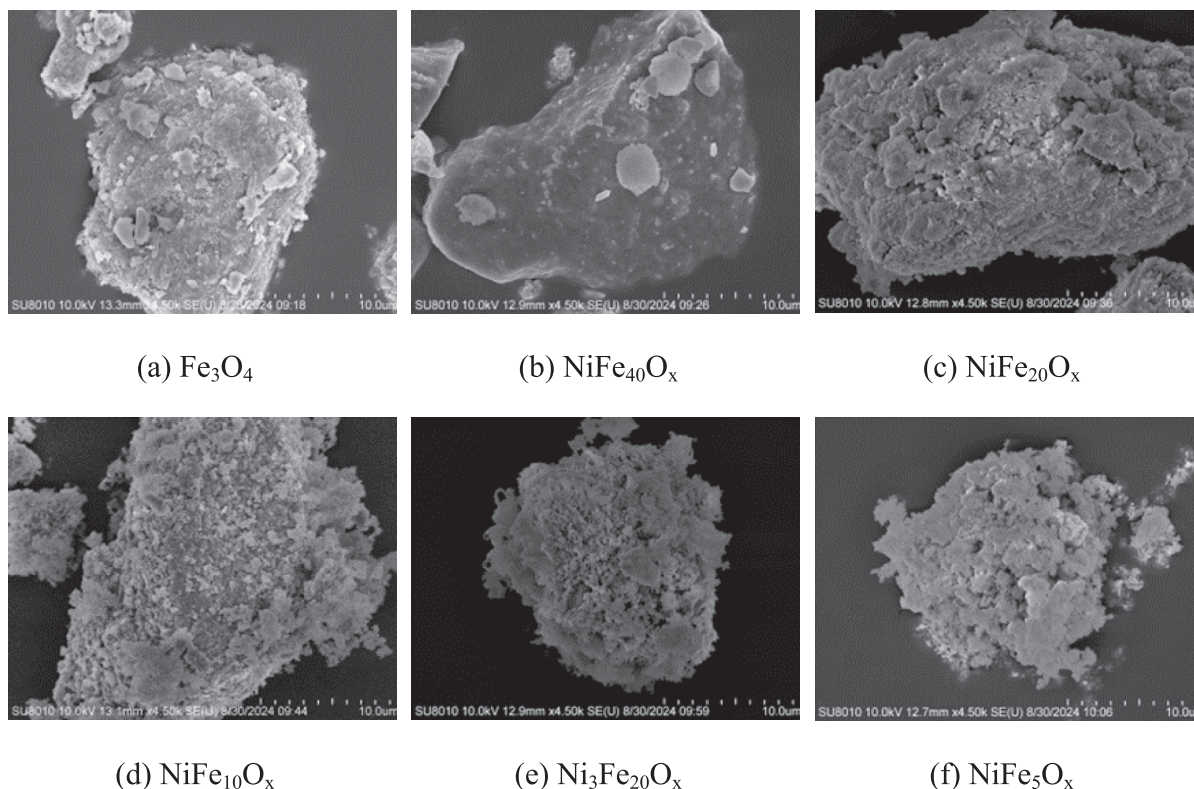


Fig. 2. SEM images of different oxygen carriers.

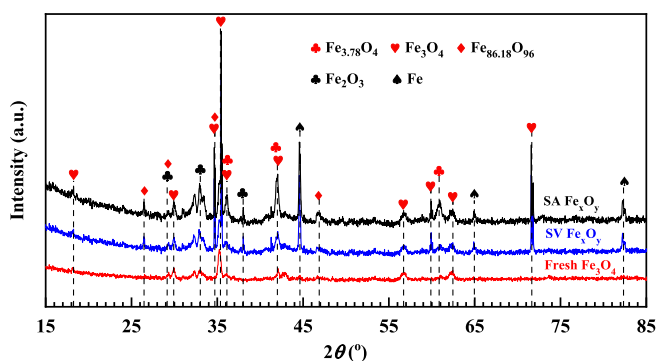
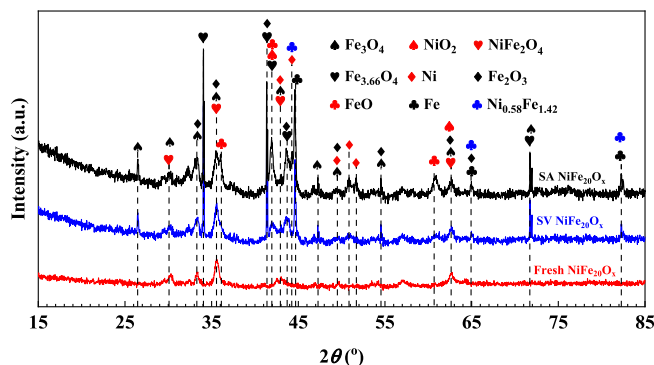
(a)  $\text{Fe}_3\text{O}_4$ (b)  $\text{NiFe}_{20}\text{O}_x$ 

Fig. 3. XRD spectra of fresh and spent oxygen carriers (SV: collected after vacuum stage, SA: collected after air introduction stage).

### 3.2. Effect of nickel loading molar ratio

The molar ratio of Ni loaded onto the oxygen carriers significantly influenced the experimental process. It notably affected the heating rate by altering the dielectric loss capacity and microwave absorption ability [35], and it also impacted the reactivity of the oxygen carriers by increasing the number of active sites, which promotes the cracking of plastic volatiles [36]. Therefore, it was essential to detail the effect of the Ni/Fe molar ratio on gas yield, gas components, and carbon conversion efficiency. A dedicated study was conducted on the chemical looping gasification of PP using  $\text{Ni}_a\text{Fe}_b\text{O}_x$  as the oxygen carrier with varying Ni/Fe molar ratios (0, 1:40, 1:20, 1:10, 3:20, and 1:5) to elucidate the effect of its influence on gas production. The experimental temperature was fixed at 900 °C, as higher temperatures favor gas production [31]. Microwave power was set at 890 W based on preliminary experiments. The results are presented in Fig. 4.

#### 3.2.1. Microwave heating performances

Fig. 4 (a) shows the microwave heating performances of the feedstocks using  $\text{Ni}_a\text{Fe}_b\text{O}_x$  as oxygen carrier with different Ni/Fe molar ratios (0, 1:40, 1:20, 1:10, 3:20, and 1:5). To enhance thermal efficiency, SiC was added to each group to improve the heating rate. The temperature of the feedstocks increased from room temperature to 900 °C in 1.5–2.5 min, with heating rates ranging from 360 to 600 °C/min.  $\text{Fe}_3\text{O}_4$  achieved the lowest heating rate, while  $\text{Ni}_3\text{Fe}_{20}\text{O}_x$  achieved the highest.  $\text{Ni}_a\text{Fe}_b\text{O}_x$  demonstrated higher heating rate than  $\text{Fe}_3\text{O}_4$ , attributed to the mixture of the two metals promoting the generation of microwave plasma with increased heating efficiency [37]. In addition, transition metal oxides can generate defects and oxygen vacancies at high temperatures, which acted as dipole centers to induce dipole polarization relaxation [35]. The incorporation of varying quantities of Ni onto  $\text{Fe}_3\text{O}_4$  changed its physical characteristics, thereby altering the dielectric loss capacity and affecting the microwave absorption ability. Due to the poor microwave absorption ability of PP, its temperature was initially lower compared to SiC

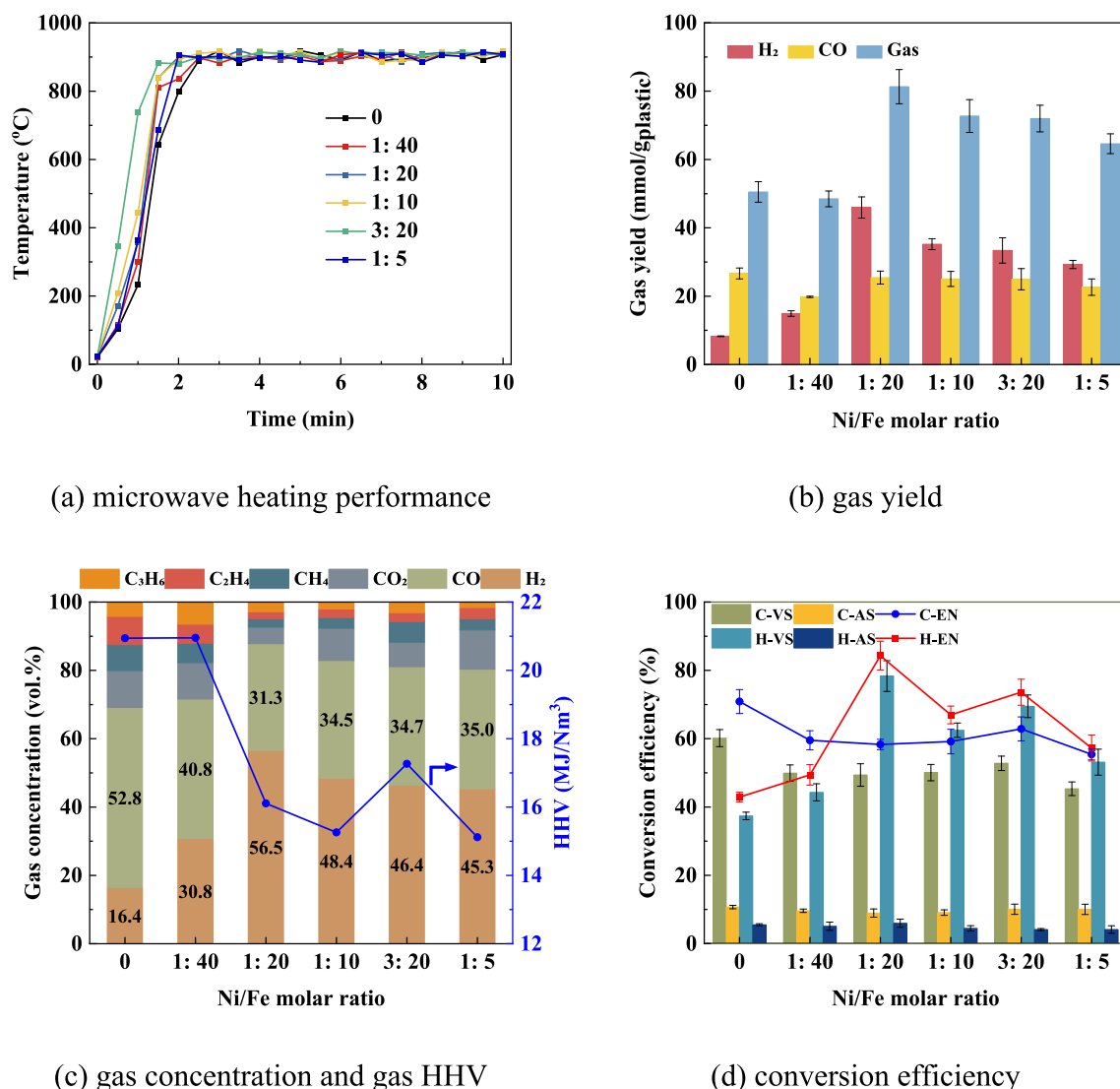


Fig. 4. Experimental results obtained from microwave-assisted chemical looping gasification of PP over  $\text{Ni}_a\text{Fe}_b\text{O}_x$  with different Ni/Fe molar ratios (VS: vacuum stage, AS: air introduction stage, EN: sum of vacuum stage and air introduction stage).

and the oxygen carrier, but it increased rapidly due to the heating from SiC and the oxygen carrier. This resulted in a temperature gradient from the oxygen carrier to the PP, aligning with the mass transfer direction, which was beneficial for plastic degradation, as demonstrated by a previous study [38].

### 3.2.2. Products distribution

Fig. 4 (b) shows the  $\text{H}_2$  yield, CO yield, and gas yield obtained from the microwave-assisted chemical looping gasification of PP using  $\text{Ni}_a\text{Fe}_b\text{O}_x$  as the oxygen carrier with different Ni/Fe molar ratios. The oil

yield is shown in Table 1. The gas yield and oil yield varied within the ranges of 48.5–81.3 mmol/g<sub>pp</sub> and 23.3–41.9 wt.%, respectively. The  $\text{H}_2$  yield and CO yield varied within the ranges of 8.3–46.0 mmol/g<sub>pp</sub> and 17.4–26.6 mmol/g<sub>pp</sub>, respectively. In the absence of Ni incorporation, a gas yield of 50.5 mmol/g<sub>pp</sub> and an oil yield of 26.7 wt.% were obtained. At a Ni/Fe molar ratio of 1:40, the gas yield decreased to 48.5 mmol/g<sub>pp</sub> while the oil yield increased to 41.9 wt.%. The incorporation of Ni increased the  $\text{H}_2$  yield from 8.3 to 14.9 mmol/g<sub>pp</sub>, as it increased the active sites and the activity of the oxygen carrier for  $\text{H}_2$  production [39], which has been confirmed by many previous studies [40]. Although  $\text{H}_2$  yield remained low, the increase in  $\text{H}_2$  yield raised the unsaturation degree of the residues, thereby improving the oil yield and decreasing the CO yield. As the Ni/Fe ratio increased to 1:20, gas yield and  $\text{H}_2$  yield increased significantly from 48.5 to 81.3 mmol/g<sub>pp</sub> and from 14.9 to 46.0 mmol/g<sub>pp</sub>, respectively. This was attributed to: (a) increased heating rate compared to  $\text{NiFe}_{40}\text{O}_x$ , promoting cleavage of C-H bonds and C-C bonds, and the yield of small molecules [32], and (b) more Ni providing additional active sites for C-H bond cleavage [36]. The lowest oil yield was due to the significant increase in  $\text{H}_2$  yield, which promoted the generation of more free carbons [41], thereby reacting with the oxygen carrier and increasing the CO yield from 19.8 to 25.4 mmol/g<sub>pp</sub>. With higher Ni molar ratios, both  $\text{H}_2$  and gas yields decreased, likely due

Table 1

Oil yields of microwave-assisted chemical looping gasification of plastics at different experimental parameters.

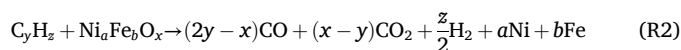
Ni/Fe molar ratio	Oil yield (wt.%)	Microwave power (W)	Oil yield (wt.%)	Plastic type	Oil yield (wt.%)
0	26.7	590	40.7	LDPE	25.2
1: 40	41.9	690	36.5	HDPE	29.3
1: 20	23.3	790	32.0	PS	70.0
1: 10	31.5	890	23.3	PP	23.3
3: 20	32.1	990	42.9	MP	44.7
1: 5	33.7	/	/	/	/

to excess Ni obstructing the active sites on the oxygen carrier [42], as observed in the SEM images in Fig. 2, thereby inhibiting the catalytic activity of  $\text{Fe}_3\text{O}_4$ . As a result of the reduction in  $\text{H}_2$  yield and free carbon, the CO yield decreased slightly. Due to the addition of the oxygen carrier, the gas yield (81.3 mmol/g<sub>PP</sub>) and  $\text{H}_2$  yield (46.0 mmol/g<sub>PP</sub>) improved significantly compared with the microwave pyrolysis of PP at 900 °C, which produced 21.8 mmol/g<sub>PP</sub> gas and 1.1 mmol/g<sub>PP</sub>  $\text{H}_2$  [43]. However, the obtained gas yield and  $\text{H}_2$  yield were lower than those from chemical looping gasification of PP with steam introduced (gas yield 129.90 mmol/g<sub>PP</sub>,  $\text{H}_2$  yield 95.23 mmol/g<sub>PP</sub>) [7], as steam significantly enhanced the gas production while reducing oil yield ( $\text{C}_y\text{H}_z + y\text{H}_2\text{O} \rightarrow y\text{CO} + (y + z/2)\text{H}_2$ ,  $\text{C} + \text{H}_2\text{O} \rightarrow \text{CO} + \text{H}_2$ ,  $\text{CO} + \text{H}_2\text{O} \rightarrow \text{CO}_2 + \text{H}_2$ , and  $3\text{Fe} + 4\text{H}_2\text{O} \rightarrow \text{Fe}_3\text{O}_4 + 4\text{H}_2$ ) [44]. Despite the gas yield was lower in this study, the absence of steam simplified the experimental system and reduced energy consumption. Additionally, steam was typically introduced with the inert carrier gas to ensure uniform distribution [45], which diluted the produced gas, thereby reducing its quality and HHV values.

### 3.2.3. Gas compositions

Fig. 4 (c) shows the gas components and gas HHV obtained from the chemical looping gasification of PP using  $\text{Ni}_a\text{Fe}_b\text{O}_x$  as the oxygen carrier with different Ni/Fe molar ratios. The volume concentrations of  $\text{H}_2$ , CO,  $\text{CO}_2$ ,  $\text{CH}_4$ ,  $\text{C}_2\text{H}_4$ , and  $\text{C}_3\text{H}_6$  varied within the ranges of 16.4–56.5%, 31.3–52.8%, 4.8–10.8%, 2.4–7.6%, 2.1–8.3%, and 1.6–6.5%, respectively. The main components of the gas were  $\text{H}_2$  and CO, attributed to the addition of the oxygen carrier. With the incorporation of Ni, the  $\text{H}_2$  concentration increased significantly, resulting from the enhancement of C-H bond breakage by Ni [36]. As the ratio of Ni increased, the  $\text{H}_2$  concentration initially increased and then decreased, with the highest  $\text{H}_2$  concentration obtained at a Ni/Fe molar ratio of 1:20. The increase in the loaded Ni ratio led to an increase in active sites; however, excessive Ni aggregated and blocked the active sites of  $\text{Fe}_3\text{O}_4$  [42]. Without the incorporation of Ni, the CO concentration was the highest, due to the reactions of hydrocarbon volatiles with the oxygen carrier (R2). Additionally, fewer H atoms were released, resulting in the highest concentrations of  $\text{CH}_4$  and  $\text{C}_2\text{H}_4$ . As shown in Fig. 4 (c), the gas HHV ranged from 15.1 to 21.0 MJ/Nm<sup>3</sup>. The lowest gas HHV was obtained with  $\text{NiFe}_5\text{O}_x$ , due to the high concentration of  $\text{CO}_2$ . The highest HHV was obtained with the incorporation of  $\text{NiFe}_{40}\text{O}_x$ , due to the highest concentrations of  $\text{C}_3\text{H}_6$ , which had high heating values [46]. The heating value of gas obtained in this study was higher than that reported in most studies on the gasification of plastics (9.5–15.46 MJ/Nm<sup>3</sup>), even after removing inert carrier gases in gas like  $\text{N}_2$  or Ar [7,47].

Fig. 4 (d) shows the carbon and hydrogen conversion efficiencies at the vacuum stage, air introduction stage, and throughout the entire experiments. The carbon and hydrogen conversion efficiencies for the entire experiments varied within the ranges of 55.4–70.8% and 42.9–84.3%, respectively. During the vacuum stage, the carbon and hydrogen conversion efficiencies varied within the ranges of 45.3–60.1% and 37.4–78.3%, respectively. It was suggested that PP mainly decomposed during the vacuum stage, and the vacuum conditions in this stage led to a high oil yield [13]. During the vacuum stage, as the temperature increased to approximately 350–450 °C [48], large molecules broke down into  $\text{H}_2$  and smaller hydrocarbons (R1) [49]. Then some smaller hydrocarbons reacted with the lattice oxygen in  $\text{Ni}_a\text{Fe}_b\text{O}_x$ , generating CO,  $\text{CO}_2$ , and  $\text{H}_2$  (R2), and some heavy hydrocarbons were purged out of the reactor and condensed into liquid oil, leading to lower carbon conversion efficiency compared to chemical looping gasification of PP with steam introduction (70.8% vs 80.2%) [7]. The Fe-Ni alloy (as shown by  $\text{NiFe}_2\text{O}_4$  in Fig. 3 (b)) could catalyze deep cracking of hydrocarbons, leading to the formation of deposited carbon and  $\text{H}_2$  [50]. In the air introduction stage, the deposited carbon reacted with the inlet oxygen, generating CO (R4) and  $\text{CO}_2$  (R5), and Ni and Fe reacted with oxygen to produce NiO (R6), FeO (R7) and  $\text{Fe}_2\text{O}_3$  (R8).



### 3.3. Effect of microwave power

Microwave power influenced the chemical looping gasification process by altering the heating rate and microwave density [15], thereby affecting gas production. Therefore, it was necessary to detail the effect of microwave power on gas production. Experiments were conducted under microwave powers of 590 W, 690 W, 790 W, 890 W, and 990 W separately to optimize the microwave power.  $\text{NiFe}_{20}\text{O}_x$  was used as the oxygen carrier due to its best gas yield and  $\text{H}_2$  yield based on Section 3.2. The obtained results are shown in Fig. 5.

#### 3.3.1. Microwave heating performances

Fig. 5 (a) shows the microwave heating performances of the feedstocks using  $\text{NiFe}_{20}\text{O}_x$  as oxygen carrier under different microwave powers (590 W, 690 W, 790 W, 890 W, and 990 W). The temperature of the feedstocks increased from room temperature to 900 °C in 1.5–8 min, with heating rates ranging from 112.5 to 600 °C/min. The microwave power of 990 W achieved the highest heating rate, while 590 W achieved the lowest. It should be noted that higher microwave power did not necessarily mean higher electricity consumption. Table 2 shows the energy consumption of the microwave oven at different microwave powers, ranging from 0.182 to 0.293 kWh. The highest energy consumption was recorded at 590 W, while the lowest was observed at 890 W. The reason could be attributed to the fact that lower microwave power required a longer time to heat the feedstocks to 900 °C, resulting in higher energy consumption.

#### 3.3.2. Products distribution

Fig. 5 (b) shows the  $\text{H}_2$  yield, CO yield, and gas yield obtained from the chemical looping gasification of PP using  $\text{NiFe}_{20}\text{O}_x$  as the oxygen carrier under different microwave powers. The oil yield is shown in Table 1. The gas yield and oil yield varied within the ranges of 41.9–81.3 mmol/g<sub>PP</sub> and 23.3–42.9 wt.%, respectively. The  $\text{H}_2$  yield and CO yield varied within the ranges of 6.2–46.0 mmol/g<sub>PP</sub> and 25.4–29.6 mmol/g<sub>PP</sub>, respectively. When the microwave power increased from 590 W to 890 W, the gas yield increased. However, when the microwave power was further increased to 990 W, the gas yield decreased while the oil yield reached its highest level. Microwave power primarily affected gas yield by influencing the heating rate. As shown in Fig. 5 (a), higher microwave power led to an increased heating rate, which was closely linked to heat and mass transfer within the plastics, as well as the residence time of the volatiles [32]. As the heating rate increased, the PP underwent rapid cracking, generating a significant amount of small volatile compounds. More of these volatiles escaped the reactor before undergoing secondary reactions, such as the recombination of small

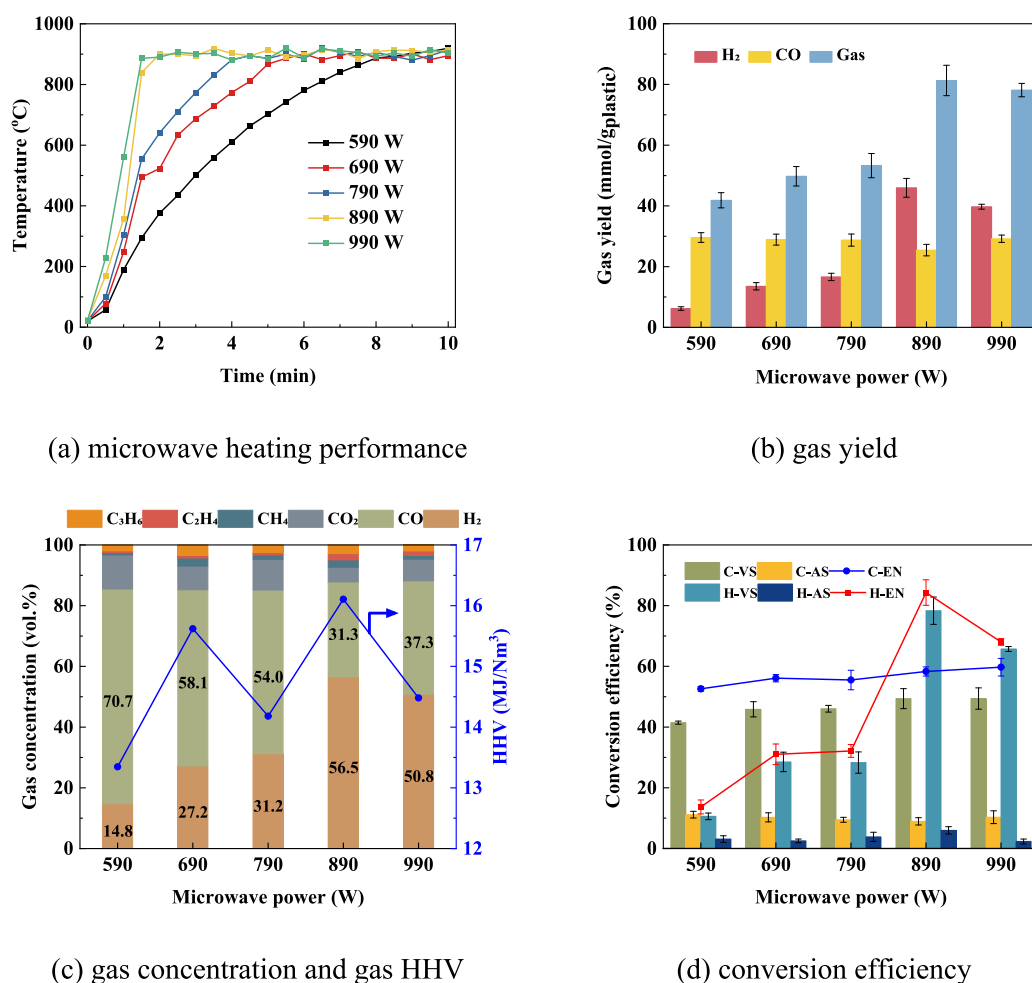


Fig. 5. Experimental results obtained from microwave-assisted chemical looping gasification of PP with different microwave powers.

Table 2

Energy consumption of microwave oven under different microwave powers.

Microwave power (W)	990	890	790	690	590
Energy consumption (kWh)	0.208	0.182	0.235	0.283	0.293

molecules [51], contributing to an increase in gas yield [16]. However, when the heating rate became excessively high, the volatiles were released too quickly, leaving insufficient time for complete cracking. Consequently, some volatiles escaped before being converted into gas products, leading to a reduction in gas yield [52]. The  $H_2$  yield initially increased and then decreased with the increase in microwave power, while the CO yield changed slightly. At 590 W, the  $H_2$  yield was only 6.2 mmol/g<sub>pp</sub>. This was attributed to the lower heating rate, which resulted in a longer residence time and promoted the recombination reactions of small molecules [51]. As the microwave power increased, the higher heating rate reduced the recombination reactions of small molecules, thereby promoting  $H_2$  production. The decrease in  $H_2$  yield at 990 W was attributed to the insufficient residence time of volatiles in the reactor, caused by the excessively high heating rate, which led to the incomplete cracking of the plastic volatiles [16].

### 3.3.3. Gas compositions

Fig. 5 (c) shows the gas components and gas HHV obtained from the chemical looping gasification of PP using  $NiFe_{20}O_x$  as the oxygen carrier under different microwave powers. The volume concentrations of  $H_2$ , CO,  $CO_2$ ,  $CH_4$ ,  $C_2H_4$ , and  $C_3H_6$  varied within the ranges of 14.8–56.5%,

31.3–70.7%, 4.8–11.2%, 0.7–2.7%, 0.7–2.1%, and 1.9–3.4%, respectively. With the increase in microwave power, the  $H_2$  concentration first increased then decreased, while the CO concentration showed the conversed tendency. The reason for this phenomenon had been explained in Section 3.3.2. The highest  $H_2$  concentration and the lowest CO concentration was achieved at microwave power of 890 W. At 590 W, some of the heavy hydrocarbons flowed out at low temperatures and were challenging to react with lattice oxygen [33]. When the temperature was sufficiently high, the residues reacted with a surplus amount of the oxygen carrier, leading to the generation of large amounts of CO and  $CO_2$ . As the microwave power increased, more smaller hydrocarbons generated in the reactors at high temperatures reacted with the oxygen carrier (R2) [47], producing a similar amount of CO but less  $CO_2$  due to insufficient oxygen. The highest gas yield was observed at 890 W, resulting in the lowest CO concentration. As shown in Fig. 5 (c), the gas HHV ranged from 13.3 to 16.1 MJ/Nm<sup>3</sup>. The lowest gas HHV was obtained at 590 W due to the highest concentration of  $CO_2$ . The highest HHV was obtained at 890 W due to the lowest concentration of  $CO_2$ .

Fig. 5 (d) shows the carbon and hydrogen conversion efficiencies at the vacuum stage, air introduction stage, and throughout the entire experiment. The carbon and hydrogen conversion efficiencies for the entire experiment varied within the ranges of 52.6–59.7% and 13.7–84.3%, respectively. During the vacuum stage, the carbon and hydrogen conversion efficiencies varied within the ranges of 41.5–49.4% and 10.6–65.7%, respectively. The carbon conversion efficiency decreased with the decrease in microwave power, due to the lower heating rate causing the breakdown of PP at lower temperatures, releasing more heavy hydrocarbons which then condensed to liquid oil



(R1) [53]. The hydrogen conversion efficiency was closely related to  $H_2$  production due to the lower production of  $CH_4$ ,  $C_2H_4$ , and  $C_3H_6$ , which could react with the oxygen carrier (R2), and the highest hydrogen conversion was achieved at 890 W.

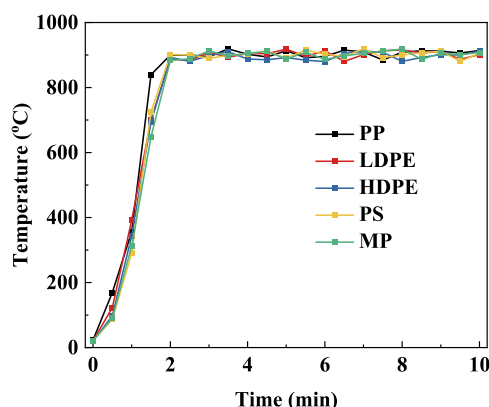
### 3.4. Effect of plastic types

$NiFe_{20}O_x$  was recognized as the optimal oxygen carrier, and 890 W was found to be the most effective microwave power for maximizing gas yield. These parameters were utilized to investigate the chemical looping gasification performance of various plastics. Four prevalent plastics, representing 50.4 wt.% of global plastic production, including PP, HDPE, LDPE, and PS, were selected to examine their microwave-assisted chemical looping gasification performances [1]. LDPE and HDPE were derived from ethylene; however, LDPE had an irregular structure with numerous side chains, while HDPE had an almost linear structure with few side chains [54]. PP, synthesized from propylene, had a relatively linear structure with a methyl group ( $-CH_3$ ) on each monomer. PS featured a linear structure with a phenyl group ( $-C_6H_5$ ) on each monomer [55]. Additionally, mixed plastics (MP) were formulated using the global production ratios (PP: LDPE: HDPE: PS = 3.63: 2.71: 2.35: 1) to simulate real plastic waste for gas production [1]. Fig. 6 presents the results of microwave-assisted chemical looping gasification over  $NiFe_{20}O_x$  under microwave power of 890 W using different plastics.

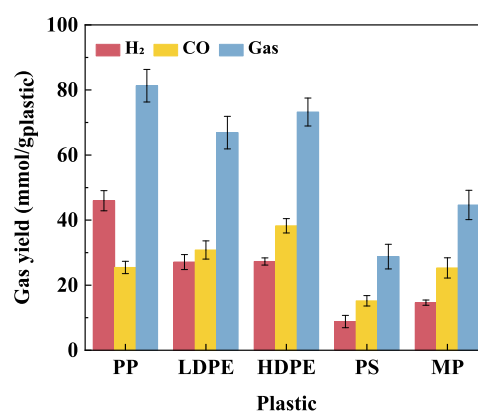
#### 3.4.1. Products distribution

Fig. 6 (a) illustrates the microwave heating performance of SiC and  $NiFe_{20}O_x$  mixed with various plastics under a microwave power of 890 W. The heating rates of the feedstocks were primarily determined by SiC and  $NiFe_{20}O_x$ , showing minimal variation based on the type of plastic, with the heating rates remaining around  $450\text{ }^{\circ}\text{C}/\text{min}$  in 2 min.

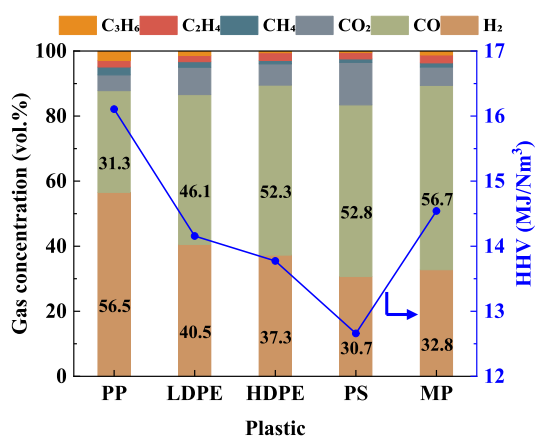
Fig. 6 (b) presents the  $H_2$  yield, CO yield, and gas yield obtained from the chemical looping gasification of different plastics using  $NiFe_{20}O_x$  as the oxygen carrier under a microwave power of 890 W. The oil yield is detailed in Table 1. The gas yield and oil yield varied within the ranges of 28.8–81.3 mmol/g<sub>Plastic</sub> and 23.3–70.0 wt.%, respectively.  $H_2$  yield and CO yield varied within the ranges of 8.8–46.0 mmol/g<sub>Plastic</sub> and 15.2–38.3 mmol/g<sub>Plastic</sub>, respectively. PS obtained the lowest gas yield,  $H_2$  yield and CO yield, while obtained the highest oil yield. This was attributed to that PS had about 74 wt.% phenyl group ( $C_6H_5$ -) which was hard to decompose (R1) resulting in the higher oil yield. Previous studies reported a gas yield of only 24.4 wt.% from PS pyrolysis at  $900\text{ }^{\circ}\text{C}$  [56]. LDPE and HDPE showed similar  $H_2$  yields, while HDPE exhibited a higher CO yield. This was attributed to HDPE's linear structure, which consisted of shorter and more regular molecular chains, requiring higher temperatures or longer reaction times for complete cracking [57]. Under these conditions, the carbonaceous fragments produced are more likely to oxidize into CO at the elevated temperature of  $900\text{ }^{\circ}\text{C}$ . In contrast, PP, with its lower crystallinity and more regular side chains, is more susceptible to thermal decomposition, resulting in its higher gas and  $H_2$  yields [58]. The synergistic effect of MP was not conducive to gas



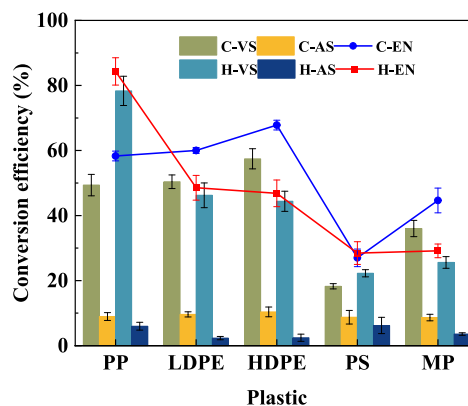
(a) microwave heating performance



(b) gas yield



(c) gas concentration and gas HHV



(d) conversion efficiency

Fig. 6. Experimental results obtained from microwave-assisted chemical looping gasification of different plastics.

production. This was evident as the gas yield (44.7 mmol/g<sub>plastic</sub>) significantly decreased compared to the average value (70.0 mmol/g<sub>plastic</sub>) of the different plastics, while the oil yield (44.7 wt.%) significantly increased compared to the weighted average value (30.1 wt.%). This was attributed to secondary reactions occurring in the generated vapors between the different plastics [53], resulting in a higher oil yield. These average values were calculated based on the weight percentages of different plastics in the MP.

### 3.4.2. Gas compositions

Fig. 6 (c) shows the gas components and gas HHV obtained from the chemical looping gasification of different plastics using NiFe<sub>20</sub>O<sub>x</sub> as the oxygen carrier under a microwave power of 890 W. The volume concentrations of H<sub>2</sub>, CO, CO<sub>2</sub>, CH<sub>4</sub>, C<sub>2</sub>H<sub>4</sub>, and C<sub>3</sub>H<sub>6</sub> varied within the ranges of 30.7–56.5%, 31.3–56.7%, 4.8–13.1%, 1.1–2.4%, 2.0–2.4%, and 0.4–2.8%, respectively. PS showed the highest CO concentration and the lowest H<sub>2</sub> concentration among different plastics, attributed to the difficulty in breaking benzene rings [56]. The carbon and hydrogen atoms in the gas mainly originated from the ethylene side chains of styrene, which contained fewer hydrogen atoms, resulting in the lowest H<sub>2</sub> concentration. HDPE and LDPE had similar H<sub>2</sub> concentrations, while HDPE showed a higher CO concentration. MP exhibited the highest CO concentration, which was due to the largest amounts of heavy molecules flowing out and the residues reacting with the surplus oxygen carrier (R2), leading to the highest CO concentration. As shown in Fig. 6 (c), the gas HHV ranged from 12.7 to 16.1 MJ/Nm<sup>3</sup>. The lowest gas HHV was observed with PS, due to its highest CO<sub>2</sub> concentration (13.1 vol.%), while PP showed the highest gas HHV with the lowest CO<sub>2</sub> concentration (4.9 vol.%).

Fig. 6 (d) shows the carbon and hydrogen conversion efficiencies during the vacuum stage, air introduction stage, and throughout the entire experiment. The carbon and hydrogen conversion efficiencies for the entire experiment varied within the ranges of 27.0–67.8% and 28.5–84.3%, respectively. During the vacuum stage, the carbon and hydrogen conversion efficiencies varied within the ranges of 18.3–57.4% and 22.3–78.3%, respectively. PS achieved the lowest carbon and hydrogen conversion efficiencies, due to its highest oil yield (70 wt.%). HDPE showed higher carbon conversion efficiency and similar hydrogen conversion efficiency compared to LDPE, due to its more regular molecular chains, which required higher temperatures or longer reaction times for complete cracking [57], making it more likely to produce CO.

### 3.5. Result comparisons

Table 3 presents the experimental results for gas production from the pyrolysis and gasification of PP, along with the findings of this study. The optimal gas yield in this study was 81.3 mmol/g<sub>pp</sub> (1.15 g/g<sub>pp</sub>), achieved using NiFe<sub>20</sub>O<sub>x</sub> as the oxygen carrier at a microwave power of 890 W and a reaction temperature of 900 °C. In comparison to electrical air gasification of PP, the gas yield in this study was lower (1.15 g/g<sub>pp</sub> vs. 1.66 g/g<sub>pp</sub>) [59], which can be attributed to the slower release rate of lattice oxygen [60]. However, the H<sub>2</sub> concentration in this study was significantly higher (56.5 vol.% vs. 11.2 vol.%) due to the catalytic effect of NiFe<sub>20</sub>O<sub>x</sub>. In air gasification, the introduction of air diluted the

generated gas, leading to a lower heating value of 7.92 MJ/Nm<sup>3</sup> compared to 16.1 MJ/Nm<sup>3</sup> in this study [59]. Similarly, microwave-assisted gasification of PP produced a lower H<sub>2</sub> concentration and heating value [61]. The H<sub>2</sub> concentration observed in this study (56.5 vol.%) was slightly lower than that reported for PP pyrolysis (59.0 vol.%), likely due to the use of a more complex catalyst in the pyrolysis process and the oxidation of lattice oxygen in this study [34]. Nevertheless, the gas yield in this study was significantly higher, owing to the oxidation of plastic volatiles, which enhanced H<sub>2</sub> production. In microwave pyrolysis of PP without a catalyst, both the gas yield and H<sub>2</sub> concentration were lower than in this study [62].

## 4. Conclusions

In this study, microwave-assisted heating coupled with chemical looping gasification of plastics at 900 °C were investigated, detailing the effects of Ni/Fe molar ratio of Ni<sub>a</sub>Fe<sub>b</sub>O<sub>x</sub> oxygen carrier, microwave power, and types of plastics on gas production. Investigations first screened Ni/Fe molar ratios (0, 1:40, 1:20, 1:10, 3:20, and 1:5) in the Ni<sub>a</sub>Fe<sub>b</sub>O<sub>x</sub> oxygen carrier. NiFe<sub>20</sub>O<sub>x</sub> emerged as the optimal oxygen carrier, achieving the highest gas yield of 81.3 mmol/g<sub>pp</sub> and an H<sub>2</sub> yield of 46.0 mmol/g<sub>pp</sub>, with a hydrogen conversion efficiency of up to 84.3 %. Using Fe<sub>3</sub>O<sub>4</sub> as the oxygen carrier, the highest gas HHV of 21 MJ/Nm<sup>3</sup> and the highest carbon conversion efficiency of 70.9 % were obtained.

Subsequently, the effect of microwave powers (590 W, 690 W, 790 W, 890 W, and 990 W) on gas production was examined. The gas yield first increased then decreased with increasing microwave power, identifying 890 W as the optimal power. The average heating rate ranged from 112.5 to 600 °C/min across the different microwave powers, with the electrical power consumption of the microwave oven varying between 0.182 kWh and 0.293 kWh. Notably, the microwave power of 890 W resulted in the lowest electrical power consumption.

Using the optimal microwave power of 890 W, the chemical looping gasification performances of different plastics over NiFe<sub>20</sub>O<sub>x</sub> were investigated. PP showed the highest gas yield and H<sub>2</sub> yield, while HDPE exhibited the highest carbon conversion efficiency of 67.8 % and the highest CO yield of 38.3 mmol/g<sub>plastic</sub>. However, the synergistic effect of MP was not conducive to gas production. This was attributed to secondary reactions occurring in the generated vapors between different plastics, resulting in a high oil yield.

### CRedit authorship contribution statement

**Wenming Fu:** Writing – original draft. **Yanling Zhang:** Supervision, Funding acquisition. **Weitao Cao:** Validation. **Wenke Zhao:** Validation. **Bingxi Li:** Supervision.

### Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

**Table 3**

Gas production from thermochemical conversion of PP.

Methods	Main conditions	Gas yield (g/g <sub>pp</sub> )	Compositions (vol.%)					Ref
			H <sub>2</sub>	CH <sub>4</sub>	CO	CO <sub>2</sub>	others	
Microwave-assisted chemical looping	890 W, 900 °C, NiFe <sub>20</sub> O <sub>x</sub>	1.15	56.5	2.4	31.3	4.8	7.4	This study
Electrical air gasification	703 °C	1.66	11.2	11.4	43.3	26.1	8.0	[59]
Microwave-assisted air gasification	800 W, 900 °C, SiC	0.86	7.2	5.4	25.7	9.8	51.9	[61]
Electrical pyrolysis	900 °C, NiCo/ZSM-5	0.65	59.0	13.5	/	/	27.5	[34]
Microwave-assisted pyrolysis	1000 W, 900 °C, SiC	0.71	11.2	12.0	/	/	76.8	[62]

## Data availability

Data will be made available on request.

## Acknowledgements

This work was financially supported by the National Natural Science Foundation of China (52076049), Heilongjiang Province “Double First-class” Discipline Collaborative Innovation Achievement Project (LJGXCG2023-080), and Heilongjiang Provincial Key R&D Program (2023ZX02C05).

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