ABSTRACT: In order to explore an efficient and clean conversion way of Ningdong coal, the chemical-looping reaction of Ningdong coal was conducted in a laboratory-scale fluidized-bed reactor using manganese ore as the oxygen carrier (OC), and the reaction characteristics were investigated in combination by thermogravimetric analysis. Experiments included the investigation of the reaction of the manganese ore with 5 vol % H2 and with coal powder at 900 °C, the catalytic and OC effect of manganese ore on coal gasification, the reaction efficiency and reaction products under different mass ratios of OC to coal (O/C), and the effect of multiple cycles on the reaction performance of the manganese ore. At 900 °C, the OC exhibited higher reactivity with coal syngas, significantly reducing the time of coal gasification (∼48% reduction). The manganese ore itself contained alkali metals, exhibiting a certain capacity for sulfur fixation, but the sulfur fixation was gradually weakened during multicycle experiments. The manganese ore did not exhibit any inhibition effect on NOx emission. From the wear rate of the manganese ore during fluidized reaction, its service life could be deduced to be about 150 h. Hence, it is necessary to improve the strength of the manganese ore OC.

INTRODUCTION

Chemical-looping combustion (CLC) technology has attracted considerable attention because it can realize graded utilization of energy and carbon dioxide capture.1−3 Thus far, the research of chemical-looping technology has extended from gas-fuel combustion to solid-fuel combustion, liquid-fuel combustion, gasification, reforming, and other fields. The main principle of CLC involves the circulation of an oxygen carrier (OC) between the fuel and air reactors, followed by the transfer of the lattice oxygen and heat needed for the reaction (Figure 1). Compared to the normal combustion of the fuel burnt in air, CLC can produce high-purity CO2 without N2 in flue gas after the condensation of vapor into water, which exhibits environmental significance.4,5 Thus, several researchers have mainly focused on the CLC/chemical-looping gasification (CLG) of coal6 because of its inherent characteristics of low NOx emissions and CO2 separation. Studies have revealed that the CLC of coal comprises four processes, namely, coal pyrolysis, char gasification, reactions between the OC and pyrolysis gas, and reactions between the OC and coal syngas.
The performance and cost of the OC, which are the core of CLC, directly determine the efficiency and economy of CLC. Typically, metal oxides such as Fe₂O₃, Mn₃O₄, NiO, and CuO are used as active OC components, and an inert component is also present to improve the physical properties of the compound, such as specific area and stability. The typical inert materials include Al₂O₃, SiO₂, TiO₂, ZrO₂, MgAl₂O₄, and so forth. An Mn-based OC, which is considered to be a potential OC, exhibits high activity, good high-temperature resistance, oxygen loading, cost-effectiveness, and relatively eclectic properties. Compared with the expensive artificially prepared OCs, low-cost natural ore OCs have attracted more and more attention. For example, active OC components but also natural inert ones are present in ilmenite, iron ore, manganese ore, and bauxite waste, which are economical and environmentally friendly; however, their performance needs to be improved.

Leion et al. reported that the activity of ilmenite increased after multiple redox cycles and finally approximated to the artificial preparation of Fe-based OCs. Li et al. have reported that loading with K⁺ leads to the production of uniform pores inside ilmenite particles after multiple cycles, thereby considerably improving the reactivity of ilmenite. Berguerand and Lyngfelt have conducted tests that show that ilmenite does not undergo sintering and is sufficiently active for application in iG-CLC systems. Wu also reported that the activity of the iron ore remained stable during the test, with a wear rate of 0.0625%/h and an iron ore lifetime of 1600 h. Matzen et al. conducted a review on the recent use of ores as OCs in CLC technologies and concluded that ilmenite presents as the most likely candidate for use as an OC and also confirmed the potential of other ores (Mn, Cu, and Ca) in the combustion of coal and syngas fuels. Fossdal has performed studies on Mn-ore-based OCs and reported that the Mn ore exhibits a higher reaction rate with H₂ and CH₄. Leion et al. found that the manganese ore exhibits poor mechanical stability and fluidization properties. For the manganese ore as an OC, however, Mei has reported that the mechanical stability and reactivity of the manganese ore are related to the characteristics and composition of the ore.

Manganese ore, as a potential OC, has attracted wide attention in academic circles, but most of the research results in this field are based on the higher quality manganese ore in China. It is necessary to study the possibility of CLC of the conventional manganese ore in China and to discuss its related characteristics. As the important coal source in China, Ningdong coal has been focused on its clean utilization. Hence, the main aim in this paper is to evaluate whether Mn-based OCs have a good oxygen-carrying performance for efficient and clean conversion of Ningdong coal and to understand the mechanisms of emission reduction during the coal conversion process. Hence, natural manganese ore is used as the OC for typical Ningdong coal fuel. CLC experiments are performed in a thermogravimetric analyzer and a laboratory-scale fluidized-bed reactor. Furthermore, effects of the reaction temperature and different mass ratios of OC to coal (O/C) on combustion results were investigated, and the release law of major pollutants was evaluated.

## EXPERIMENTAL SECTION

### OC and Coal
Ningdong coal used in the experiment was pulverized and sieved to a powder of 0.3–0.7 mm. Table 1 summarizes the elemental analysis and industrial analysis data. The used OC comprised 40% manganese-bearing natural manganese ore from the Daji Manganese Industry, which was crushed and calcined in a muffle furnace at 900 °C under air for 4 h and then sieved to a powder of 0.1–0.3 mm. The prepared OC was analyzed using an X-ray fluorescence (XRF) spectrometer, and the results are shown in Table 2.

### Reaction Equations
The possible reaction equations for the experimental process are shown below, in which the total reaction formula between the OC and coal powder is 

$$\text{C}_x\text{H}_{2m} + (2n + m)\text{MeO}_y \rightarrow n\text{CO}_2 + m\text{H}_2\text{O} + (2n + m)\text{Me}_x\text{O}_{y-1} \quad (R1)$$

coal + water vapor = char + tar + syngas \quad (R2)

$$\text{C} + 2\text{Mn}_3\text{O}_4 = 6\text{MnO} + \text{CO}_2 \quad (R3)$$

$$\text{C} + \text{Mn}_3\text{O}_4 = 3\text{MnO} + \text{CO} \quad (R4)$$

$$\text{CH}_4 + 4\text{Mn}_3\text{O}_4 = 12\text{MnO} + \text{CO}_2 + 2\text{H}_2\text{O} \quad (R5)$$

$$\text{CO} + \text{Mn}_3\text{O}_4 = 3\text{MnO} + \text{CO}_2 \quad (R6)$$

$$\text{H}_2 + \text{Mn}_3\text{O}_4 = 3\text{MnO} + \text{H}_2\text{O} \quad (R7)$$

$$6\text{MnO} + \text{O}_2 = 2\text{Mn}_3\text{O}_4 \quad (R8)$$

### Thermogravimetric Analysis Experiments
The designed experiments could be divided into three stages. In the first stage, during the uniform increase in temperature, the thermal stability of OC was investigated to check whether this manganese ore OC exhibited characteristics of oxygen decoupling. During the second stage, the reaction characteristics of the manganese ore OC and H₂ were investigated.
During the third stage, the direct contact reaction between the manganese ore OC and coal powder was investigated. Table 3 summarizes the experimental conditions, to show four groups of experiments. The thermogravimetric analyzer used here is Hengen Scientific Instrument HCT-4; its sample weight range is 1–300 mg and its sensitivity is 0.1 μg. For the sample weight variation is repeatable, the analysis is of less error.

**Fluidized-Bed Reactor Experiments.** Figure 2 shows the schematic diagram of the fluidized-bed reactor system. The inner diameter and height of the reactor were 32 and 800 mm, respectively, and the air distribution plate was 100 mm from the bottom of the reactor. The flow of gases (such as air and N₂) was controlled using a float flowmeter.

Table 3. Conditions for TGA Experiments

| exp. | temperature (°C) | heating rate (°C/min) | component | atmosphere |
|------|------------------|-----------------------|-----------|------------|
| 1    | 20–1000          | 10                    | manganese ore | N₂         |
| 2    | 20–1000          | 10                    | manganese ore & coal | N₂         |
| 3    | 20–1000          | 10                    | manganese ore | N₂         |

Table 4. Conditions for Fluidized-Bed Reactor Experiments

| exp. | temperature (°C) | coal (g) | bed material | fluidized gas | vol flow (L/min) |
|------|------------------|---------|--------------|---------------|-----------------|
| 4    | 900              | 0.1     | quartz sand  | N₂            | 2               |
| 5    | 900              | 0.1     | manganese ore & quartz sand | N₂ | 2               |
| 6  | 900              | 0.1     | manganese ore | 6.44 | N₂ & air | 2               |

“Multicycle experiment; Δ: the OC mass is added from 0 g in the first experiment to 10 g by adding 2 g each time.”

Figure 2. Schematic of the experimental setup used in this investigation.

**Data Analysis.** The oxygen demand for the complete combustion of per kg coal and theoretical oxygen release of the manganese ore OC can be calculated as eqs 1–3.

\[
V_{\text{air-coal}} = 0.0889 \times (\omega_C + 0.375 \times \omega_H) + 0.265 \times \omega_H - 0.0333 \times \omega_O
\]  
\[
M_{O_2-coal} = \frac{1000 \times V_{\text{air-coal}}}{22.4} \times 21\%
\]  
\[
M_{O_2-OC} = \frac{m_{\text{OC}}w_{\text{OC}}}{2 \times M_{\text{MnO}} + \frac{m_{\text{OC}}w_{\text{FeO}}}{6} \times M_{\text{FeO}}}
\]

Here, the numerical coefficients in eq 1 are derived from the oxidation reaction formulae of combustible elements of C, S, and H. After the consumed oxygen is calculated, the air volume is converted for per kg coal combustion, \((\text{m}^3/\text{kg})\), \(\omega_i\) is the mass fraction (\(i = \text{C, S, H, O in coal, and Mn}_3\text{O}_4, \text{and Fe}_2\text{O}_3\) in OC), \(m_{\text{OC}}\) is the OC mass, (g), and \(M\) is the molar mass of the corresponding oxide (g/mol).

Furthermore, set \(\rho_n\) represents the gaseous mass concentration at the outlet of the fluidized-bed reactor in the CLC experiment (mg/m³), and the total amount of a single component gas can be calculated as eq 4, \(m_{\text{gas}}\)

\[
m_{\text{gas}} = \frac{V_{\text{N}_2} \int_{t_1}^{t_2} \rho(i,t) \, dt}{1000}
\]

Here, \(V_{\text{N}_2}\) is the carrier gas flow rate, \((\text{m}^3/\text{s})\), \(\rho(i,t)\) is the component ratio of \(i\) gas at the \(t\) moment \((i = \text{CO, H}_2, \text{CH}_4, \text{SO}_2, \text{NO}_x\) and so forth), and \(t_1\) is the reaction start time. The conversion efficiency of the “i” component gas is expressed as follows

\[
a_i = \frac{m_{\text{gas}}M_i}{m_{\text{coal}}w_i} \times 100\%
\]

Here, \(M_{\text{gas}}\) is the molar mass of the “i” component gas, (g/mol), and \(m_{\text{coal}}\) is the coal mass, (g). \(w_i\) is the mass fraction of “i” element in pulverized coal, (%). Equation 5 is mainly used to calculate the CO/CO₂ conversion rate to analyze the temperature, followed by the addition of the OC through the funnel in the upper part of the furnace and maintenance of fluidization through 2 L/min of air at least for 10 min to ensure that the OC reached the reaction temperature and was completely oxidized. Finally, the flowmeter was adjusted to switch the fluidized gas to N₂, and the inert gas environment was guaranteed in the reactor. The vapor coming from the generator was loaded by N₂ and the mixture gas of N₂-vapor passed the preheater together, and then, the heated gas entered the fuel reactor. The regulation method of vapor supply is by the heating power of the vapor generator, and the vapor flow rate is calculated according to the water volume reduction in the generator.

Notably, to avoid the change in the bed material height caused by the change of OC mass in exp. 5, quartz sand must be added to ensure consistency with respect to the bed material volume (~25 mL) of each test sample. On the other hand, exp. 6 included the weighed OC according to the oxygen demand for the complete combustion of 0.1 g of coal powder.

In the process of experiments, the gas at the outlet of the fluidized-bed reactor was detected using a gas analyzer (Kane Quintox, KANE 9506).

**Table 3. Conditions for TGA Experiments**

| exp. | temperature (°C) | heating rate (°C/min) | component | atmosphere |
|------|------------------|-----------------------|-----------|------------|
| 1    | 20–1000          | 10                    | manganese ore | N₂         |
| 2    | 20–1000          | 10                    | manganese ore & coal | N₂         |
| 3    | 20–1000          | 10                    | manganese ore | N₂         |
combustion situation and the conversion rate of polluting elements (such as S and N) in coal.

In multicycle experiments, the mechanical properties of the OC were investigated, and its lifetime was evaluated as eq 6, only considering the influence of mechanical strength.

\[ t_{OC} = \frac{m_{t}}{\Delta m} (t - t_{0}) \]  
(6)

“\( m_{t} \)” is the weight of OC before the start of the exp. 6, (g).

“\( \Delta m \)” is the reduced mass after rescreening of OC after “\( t \)” hours of cycling, (g).

The total low heating value “\( Q \)” and heat release rate “\( q \)” are calculated as follows

\[ V_i = \frac{22.4 \times m_{i,\text{gas}}}{M_{i,\text{gas}}} \]  
(7)

\[ Q = \frac{12.64V_{\text{CO}} + 10.79V_{\text{H}_2} + 35.88V_{\text{CH}_4}}{\sum V_i} \]  
(8)

\[ q = \frac{Q}{t} \]  
(9)

Here, “\( V_i \)” is the volume of each gaseous component in the product gas (L), calculated from the total gas mole volume, and “\( t \)” is the reaction duration. The constants in eq 8 are the calorific value of the corresponding gas (MJ/N m³). Equations 7–9 can be utilized to evaluate the transition state of CLC to CLG, according to the variation of low-heating value.

### RESULTS AND DISCUSSION

During the thermal stability experiment, no clear weight loss of the manganese ore was observed at 1000 °C, indicative of the good stability and the absence of oxygen decoupling properties. Mei tested the oxygen decoupling properties of four kinds of manganese ores at 800, 850, and 950 °C, and no obvious weight gain and weight loss were observed,25 which is consistent with the results of this paper. Therefore, the decoupling performance is not discussed here.

**Reaction Characteristics of the Manganese Ore in H₂ Atmosphere.** Figure 3 shows the thermogravimetric analysis (TGA) curve of the manganese ore OC in an atmosphere of 5 vol % H₂/N₂. The total weight loss rate of the manganese ore is about 9.03%, and the clear weight loss achieved two peaks.

First, when the temperature reaches 435.7 °C, the manganese ore begins to lose weight and the weight loss rate reaches the maximum at 627.2 °C, and DTG is ~0.24%/min, corresponding to the Mn-based OC react with H₂. Then, the Fe-based component in the ore OC begins to react with H₂ at 791.5 °C; hence, the second peak appears in the DTA curve, which is smaller than before, only ~0.21%/min. These characteristics indicate that H₂ reacts with the lattice oxygen very rapidly (as shown in R7), easily with the Mn-based OC, and then with the Fe-based OC at higher temperatures.

**Reaction Characteristics of the Manganese Ore and Coal.** In exp. 3, the direct reaction of the manganese ore with coal powder was examined by TGA. As in Figure 4, the weight loss curve of the manganese ore OC was divided into three stages: the first weight loss peak at 91.9 °C, corresponding to the evaporation of moisture from coal. At 263.6–567.4 °C, the weight loss rate of a mixture of manganese ore and coal powder was ~3.63% mainly because of the release of volatile matter from coal. After the transition section of 570–712 °C and at 712–1000 °C, both the Mn-based OC and Fe-based OC in the ore achieved high activity, thus achieving a weight loss peak of ~7.05% in the well-exposed mixture. Data from above indicate that this manganese ore OC exhibits a solid–solid reaction with the coal particles, as the reactions in R3 and R4.

Performing the above gas–solid reaction of the manganese ore with H₂ and the solid–solid reaction with coal powder particles, the complete reaction temperatures are below 1000 °C, which proves that this manganese ore OC has high activity and can be used in the CLC experiment of coal.

**Effect of the Mass Ratio of OC to Coal (O/C) in the CLC Reaction.** In exp. 6, the effect of different OC–coal mass ratios (O/C) on the CLC reaction was investigated. The mass of coal powder in each experiment was fixed at 0.1 g, and the OC mass is added from 0 g in first experiment to 10 g by adding 2 g each time. According to the calculation shown in eqs 1 and 2, it equals to the addition of 0.0064 mol of oxygen in each experiment. Concentrations of CO and CO₂ at the outlet of the reactor were monitored to determine the combustion situation of coal powder. Figure 5 shows the experimental results.

The curves in the Figure 5a,c represent the CO, CO₂, and hydrocarbon (HC) gas concentrations as a function of the reaction time, and Figure 5b shows the conversion rate of fixed
The concentration changes of CO and CO$_2$ were mainly divided into two stages. First, the pulverized coal was rapidly heated by the OC and quartz sand particles in the material layer, and a large amount of CO and CO$_2$ originated from volatile matter and coal pyrolysis (within the first 60 s), followed by the start of the coal char gasification reaction. With the increase in the manganese ore, the peak CO concentration and the subsequent gasification concentration decreased, while the CO$_2$ concentration increased. In terms of the carbon conversion rate, the conversion rate of C—to CO gradually decreased, and the conversion rate of C—to CO$_2$ increased, indicating that the manganese ore reacts with coal powder, as shown in R3, R4, and R6. Nevertheless, the total carbon conversion rates of C to CO and CO$_2$ slightly decreased (~11% reduction). Combined with Figure 5c, compared to the pure coal gasification with 0 g OC, the addition of the manganese ore slightly affected the peak value of HC gas release while significantly prolonged the release time of HC gas; therefore, the increase in cumulative HC release causes the total carbon conversion rates of C to CO and CO$_2$ decreased. The manganese ore exhibits certain characteristics for the reforming coal synthesis gas into HC gas.

In addition, with the increase in the manganese ore mass, the reaction time was gradually shortened, indicating that the manganese ore exhibits the function of the catalytic coal gasification rate and that the gasification time decreases by ~48% in this experiment.

To investigate the best gasification conditions, the total amounts of CO, CO$_2$, H$_2$, and CH$_4$ in tail gas under different (O/C) conditions were investigated, according to eq 7, and the results are shown in Figure 6a. From the calorific value of combustible gas (CO, H$_2$, CH$_4$) and the total volume of tail gas produced by the fluidized-bed reactor, the total low heating value under different O/C can be calculated according to eq 8, and then, the low heating value can be calculated according to eq 9 and shown in Figure 6b.

With the increase in the OC addition and combustion efficiency, the total low heating value of the tail gas gradually decreased, but the heat release rate first increased and then started to decrease when O/C ratio greater than 40, while the combustible gas produced per unit time begins to decrease. For the manganese ore provided oxygen for burning as the OC and less catalysis for coal gasification, the reaction between the manganese ore and coal begins to change from CLG to CLC. However, at the O/C ratio ranging between 20 and 40, the total low heating value of the outlet gas decreased, but the gasification time was shortened by 22.5−38.6%, and the heat release rate was greater than that of the other ranges. Hence, coal gasification is suitable under the condition of O/C ratio less than 40, combustion suitable under O/C ratio larger than 40.

**Multicycle Experiments and OC Performance Analysis.** According to eqs 1−3, lattice oxygen released from 6.44 g of the manganese ore was equal to the amount of the oxygen demand for complete coal combustion, which was used in the multicycle experiment (Figures 7 and 8).

From Figure 7, the first five cycle curves revealed that the reduction reaction between OC and coal powder is completed in 15 min and that the OCs take ~5 min for complete regeneration. The CLC experiment could realize stable cycling through the switching atmosphere system. In Figure 8, the conversion rate of fixed carbon into CO of the first 10 cycles was basically stable at 10−11.5%, and then, it exhibited an upward trend and finally at ~17% after 20 cycles. With the increase in the cycle times, the reaction time increased, indicating that the oxygen supply capacity of the OC is clearly attenuated because the lattice oxygen consumed less CO after 20 cycles, thereby leading to the increase in CO
concentration in flue gas. For this manganese ore OC, it is crucial to add new OCs to maintain the oxygen supply capacity and conversion rate after 15 cycles. The oxygen regeneration in the air reactor is always sufficient, the attenuation of oxygen supply performance of the OC is related to surface reaction characteristics, such as carbon deposition, sintering, and oxygen vacancy loss.

Pollutant Release Characteristics. To investigate the effect of different (O/C) ratios and cycle times on pollutant emission, the concentration variation of the pollutants \( \text{SO}_2, \text{NO}_x \) in exp. 5 and exp. 6 are shown in Figure 9.

Figure 9a shows the conversion ratio of the N and S content in coal to \( \text{NO}_x \) and \( \text{SO}_2 \) with different O/C ratios. Most of the sulfur is known to be converted to \( \text{SO}_2 \) via sulfur balance calculation, with no clear release of \( \text{H}_2\text{S} \) detected in several experiment groups. Therefore, it is determined that \( \text{SO}_2 \) is the main converted form of sulfur. As can be observed in (b), the peak for the \( \text{SO}_2 \) release was observed at the coal pyrolysis stage, which was then slowly released at the coal char gasification stage. With the increase in the OC mass, the conversion or release concentration of \( \text{SO}_2 \) decreased in comparison to the 0 g OC experiment, and the total amount of \( \text{SO}_2 \) released after the addition of OC decreased by \( \sim 72\% \). For the presence of a certain amount of alkali metal oxide (such as \( \text{CaO} \) and \( \text{MgO} \)) in the manganese ore, as in Table 2, the manganese ore OC exhibits certain desulfurization ability, shown in Figure 9, by alkali metal oxide reacting with \( \text{SO}_2 \) to form the corresponding sulfate. However, from (b), with the increase in the cycle times, the conversion rate of \( S \) into \( \text{SO}_2 \) increased, and the existence time of \( \text{SO}_2 \) gradually increased, indicating that the alkali metal in OC is continuously consumed and that the sulfur fixation efficiency decreases. Hence, it is necessary to supplement fresh OC to keep the desulfurization capacity.

In terms of the \( \text{NO}_x \) concentration curve, with the increase in O/C ratio, the conversion ratio of N in coal to \( \text{NO}_x \) gradually increased. Based on the reaction temperature of 900 \( ^\circ \text{C} \), thermal \( \text{NO}_x \) are not considered, only the fuel \( \text{NO}_x \) is produced by the oxidation of N in coal by lattice oxygen. As the OC mass increases, the supply of lattice oxygen increases, resulting in more \( \text{NO}_x \) generation. In 30 cycle experiments, the \( \text{NO}_x \) release curve did not exhibit a variation, and the conversion ratio of N to \( \text{NO}_x \) was stable. Therefore, the manganese ore OC does not exhibit a nitrogen fixation effect.

After 30 cycle experiments, the OC was sieved and weighed again, and the ore particles of less than 0.01 mm are considered to be completely broken to characterize the loss of OC. The weight of the OC was 5.802 g; this value is 9.907\% less than the weight (6.44 g) before the experiment. Moreover, from the eq 6 calculation, the working life of this manganese ore OC is \( \sim 150 \) h. Hence, the decrease of the manganese ore weight causes the decrease of the lattice oxygen provided, then leads to the conversion rate of fixed carbon into CO increasing during the 30 cyclic reactions in Figure 8. Therefore, in order
to ensure the normal and efficient CLC, fresh manganese ore OC must be replenished in time after 15 cyclic reactions.

**CONCLUSIONS**

In this paper, the chemical-looping reaction of the manganese ore with CO/H₂ and Ningdong coal is investigated in a laboratory-scale fluidized-bed reactor by TGA. The characteristics of pollutant release and the mechanical strength of the manganese ore are analyzed by multicycle experiments, and the reaction performance of the manganese ore is verified. The preliminary conclusions are as follows:

1. At 900 °C, the manganese ore OC exhibits better reactivity with H₂, and there is also a direct solid–solid reaction that occurred with char particles.

2. In the fluidized-bed reactor, with the increase in O/C ratio, the CO concentration in the tail gas gradually decreases, the CO₂ concentration increases, and the ratio of fixed carbon to CO₂ increases. Compared to coal gasification, when the O/C ratio is between 20 and 40, the heat release rate of fuel gas is the highest and the reaction time is shortened by 22.5–38.6%. This range is considered to be suitable for CLG.

3. SO₂ is the main product form of sulfur in the chemical-looping reaction of coal whether the O/C is lower or higher, and the presence of alkaline metal oxides in OC imparts a certain desulfurization capacity. However, with the increase in cycle times, the sulfur fixation capacity decreases. The manganese ore did not exhibit a fixed nitrogen effect, but prompted lattice oxygen to oxidize the nitrogen-containing substances in coal to produce NOₓ.

4. After multicycle experiments, the OC clearly exhibits wear, and the poor mechanical strength of the manganese ore OC needs to be further strengthened.

**AUTHOR INFORMATION**

**Corresponding Author**

Cuiping Wang — College of Electrical and Mechanical Engineering, Qingdao University, Qingdao 266071, China; College of Civil Engineering and Architecture, Shandong University of Science and Technology, Qingdao 266590, China; orcid.org/0000-0002-4249-709X; Email: wangcuiping@tsinghua.org.cn

**Authors**

Wenzheng Liang — College of Electrical and Mechanical Engineering, Qingdao University, Qingdao 266071, China

Kun Wang — College of Electrical and Mechanical Engineering, Qingdao University, Qingdao 266071, China

Shirui Yuan — College of Civil Engineering and Architecture, Shandong University of Science and Technology, Qingdao 266590, China

Qingjie Guo — State Key Laboratory of High-Efficiency Utilization of Coal and Green Chemical Engineering, Ningxia University, Yinchuan 750021, China

Xiude Hu — State Key Laboratory of High-Efficiency Utilization of Coal and Green Chemical Engineering, Ningxia University, Yinchuan 750021, China

Complete contact information is available at: https://pubs.acs.org/10.1021/acsomega.0c03464

**Notes**

The authors declare no competing financial interest.

**ACKNOWLEDGMENTS**

This work was supported by the National Natural Science Foundation of China (grant no. 51676102). The authors greatly acknowledge the support from the Foundation of State Key Laboratory of Coal Clean Utilization and Ecological
Chemical Engineering (grant no. 2018-K03) and the Key Technology Research and Development Project of Shandong Province (grant no. 2019GGX103001).

■ REFERENCES

(1) Li, Z. S.; Bao, J. H.; Sun, H. M. Research and development of coal-fueled chemical looping combustion. Proc. of CSEE 2014, 34, 5131–5139.

(2) Lyngfelt, A. Chemical-looping combustion of solid fuels-status of development. Appl. Energy 2014, 113, 1869–1873.

(3) Demirel, Y.; Matzen, M.; Winters, C.; Gao, X. Capturing and using CO2 as feedstock with chemical looping and hydrothermal technologies. Int. J. Energy Res. 2015, 39, 1011–1047.

(4) Petrakopoulou, F.; Alicia, B.; Marlene, C. Exergoeconomic and exergoenvironmental analyses of a combined cycle power plant with chemical looping technology. Int. J. Greenhouse Gas Control 2011, 5, 475–482.

(5) Jin, H.; Hong, H.; Han, T. Research progress on energy and environment system of chemical looping combustion. Chin. Sci. Bull. 2008, 53, 2994–3005.

(6) Wang, G.; Wang, S.; Luo, M. Research advances in chemical-looping combustion for solid fuels. Chem. Eng. Prog. 2010, 8, 1443–1450.

(7) Johansson, M. Screening of Oxygen-Carrier Particles Based on Iron-, Manganese, Copper- and Nickel Oxides for Use in Chemical-Looping Technologies; Chalmers University of Technology, 2007.

(8) Johansson, M.; Mattisson, T.; Lyngfelt, A. Comparison of oxygen carriers for chemical-looping combustion. Therm. Sci. 2006, 10, 93–107.

(9) Mattisson, T.; Järnklä, A.; Lyngfelt, A. Reactivity of some metal oxides supported on alumina with alternating methane and oxygen application for chemical-looping combustion. Energy Fuels 2003, 17, 643–651.

(10) Cho, P.; Mattisson, T.; Lyngfelt, A. Defluidization conditions for a fluidized-bed of iron oxide, nickel, and manganese oxide containing oxygen-carriers for chemical-looping combustion. Ind. Eng. Chem. Res. 2006, 45, 968–977.

(11) Jiang, L.; Zhao, H.; Zhang, S. Multiple-cycle reaction performance of sol-gel-derived Fe2O3/Al2O3 oxygen carrier with methane. J. Eng. Thermophys. 2011, 32, 329–332.

(12) Adánez, J.; Gayun, P.; Celaya, J. Chemical looping combustion in a 10kWt prototype using a CuO/Al2O3 oxygen carrier: Effect of operating conditions on methane combustion. Ind. Eng. Chem. Res. 2006, 45, 6075–6080.

(13) Xu, L. Synthesis and Characteristic Investigation of Cu-Based and Mn-Based Oxygen Carriers for Chemical Looping Combustion; Tsinghua University, 2016.

(14) León, H.; Mattisson, T.; Lyngfelt, A. Solid fuels in chemical-looping combustion. Int. J. Greenhouse Gas Control 2008, 2, 180–193.

(15) León, H.; Lyngfelt, A.; Johansson, M.; Jerndal, E.; Mattisson, T. The use of ilmenite as an oxygen carrier in chemical-looping combustion. Chem. Eng. Res. Des. 2008, 86, 1017–1026.

(16) Adánez, J.; Quadrat, A.; Abad, A. Ilmenite activation during consecutive redox cycles in chemical-looping combustion. Energy Fuels 2010, 24, 1402–1413.

(17) Bao, J.; Li, Z.; Cai, N. Promoting the reduction reactivity of ilmenite by introducing foreign ions in chemical looping combustion. Ind. Eng. Chem. Res. 2013, 52, 6119–6128.

(18) Bao, J.; Li, Z.; Cai, N. Reduction kinetics of foreign ion promoted ilmenite using carbon monoxide (CO) for chemical looping combustion. Ind. Eng. Chem. Res. 2013, 52, 10646–10655.

(19) Berguerand, N.; Lyngfelt, A. Design and operation of a 10 kWe (th) chemical-looping combustor for solid fuels-testing with South African coal. Fuel 2008, 87, 2713–2726.

(20) Berguerand, N.; Lyngfelt, A. The use of petroleum coke as fuel in a 10 kWt chemical-looping combustor. Int. J. Greenhouse Gas Control 2008, 2, 169–179.