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Release characteristics of mercury in chemical looping combustion of bituminous coal

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ABSTRACT

This study evaluated the release characteristics of mercury from bituminous coal in chemical looping combustion (CLC) using Australian iron ore as the oxygen carrier in a fixed bed reactor. The effects of several parameters, such as temperature in the fuel reactor (FR) and air reactor (AR), gasification medium in the FR, and reaction atmosphere in the AR, on mercury release characteristics, were investigated. The mercury speciation and release amount in the FR and AR under different conditions were further explored. The results indicate that most of the mercury in coal was released in the FR, while the rest of it was released in the AR. Hg^0 was found to be the major species in the released mercury. The results also indicate that a higher temperature in the FR led to an increase in the total mercury release amount and a decrease in Hg^0 proportion. However, a higher temperature in the AR resulted in a decrease in the total mercury release amount and Hg^0 proportion. The increase in the $\text{H}_2\text{O}/\text{CO}_2$ ratio of gasification mediums in the FR was beneficial for the increase in the total mercury release amount and Hg^0 proportion. A higher O_2 concentration in reaction atmosphere in AR had a negligible effect on the total mercury release amount, but a positive effect on Hg^0 oxidation.

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Introduction

Mercury (Hg) is considered one of the most toxic metals which can affect human health because of its volatility, bioaccumulation, and persistence in atmosphere (Mukherjee et al., 2004). Coal-fired power plants are proven as one of the major sources of global atmospheric mercury pollution (Tian et al., 2012). Although the amount of mercury in coal is relatively low (average concentration is about 0.22 mg/kg), it can be directly emitted

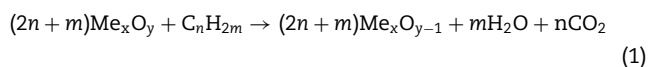
into the atmosphere with flue gas after combustion (Wang et al., 2000). Thus, mercury emission from coal-fired power plants has attracted increased attention from legislators, policymakers, and researchers (Yin et al., 2013). Numerous efforts have also been made to reduce mercury emissions from coal-fired power plants. In 2011, the US Environmental Protection Agency issued the Mercury and Air Toxics Standards to reduce the emission of mercury and other toxic pollutants from power plants. During the same period, China also issued a new air pollutant emission standard for coal-fired power plants, which specified emission concentration limits for mercury and its compounds (Sun and Zhang, 2012). Furthermore, studies on mercury in coal-fired flue gas show that it mainly exists in three forms: oxidized Hg (Hg^{2+}), particulate Hg (Hg^p), and elemental Hg (Hg^0) (Huang

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et al., 2016). Among them, Hg^0 accounts for more than 50% of total mercury in flue gas (Shah et al., 2008). Owing to its low water solubility, volatility, and inert nature, Hg^0 is hard to remove with existing air pollution control devices (Pavlish et al., 2003).

In recent years, a promising chemical looping combustion (CLC) process has been proposed, which can not only achieve a high combustion efficiency, but also a low cost of carbon capture (Shen et al., 2008, 2009; Song et al., 2012b). In CLC, the oxygen carrier is circulated between fuel reactor (FR) and air reactor (AR), transferring oxygen from air to the fuel. The oxygen carrier first reacts with fuels in FR, and then, the reduced oxygen carrier is transported into AR to be re-oxidized with air. Two reactions are involved in this process, as described in Eqs. (1) and (2) (Leion et al., 2008). The oxidation reaction is always exothermic, while the reduction reaction may be exothermic or endothermic (García-Labiano et al., 2005). Nevertheless, the sum of the enthalpies from Eqs. (1) and (2) is the same as that from conventional combustion process. In addition, since flue gas from FR is not diluted with atmospheric nitrogen, the gaseous effluent mainly consists of CO_2 and H_2O . That is, after flue gas condensation, a high concentration of CO_2 can be obtained without a high energy consumption process (Shen et al., 2008).



However, more attention has been paid to CO_2 capture and SO_x/NO_x emission in CLC process (Gu et al., 2015; Ma et al., 2018; Song et al., 2012a), and only a few studies have explored mercury release and its transformation characteristics. Mendiara et al. (2014) explored mercury release in an in-situ gasification CLC process and found temperature and coal type to have a significant influence on mercury release fraction. Considering that pyrolysis is the first step in coal combustion and gasification and plays an important role in chemical reactions (Luo et al., 2019), Ma et al. (2019) further investigated mercury release in coal pyrolysis and char gasification of CLC process and found that about 44.7% of mercury was released in the gas-phase form within the coal pyrolysis process, whereas 13.4% was released during the char gasification process. Nevertheless, mercury release characteristics in CLC process at different operating conditions are still unclear and should be investigated further.

In this paper, the release characteristics of mercury in a CLC process of bituminous coal were studied with a fixed bed reactor, which was adopted to simulate the CLC pattern. The release amount and proportion of mercury in FR and AR at different operating parameters were evaluated. Besides, gasification mediums and O_2 concentrations in FR and AR of CLC process on mercury releasing characteristics were also studied. Understanding the mercury release characteristics during CLC can be helpful in predicting mercury amount and speciation in flue gas from the reactor, which will benefit the research on mercury control.

1. Experimental section

1.1. Oxygen carrier and fuel

Australian iron ore ($\text{Fe}_2\text{O}_3/\text{Fe}_3\text{O}_4$ pellets) in the size range of 0.3–0.5 mm, provided by Nanjing Iron Steel Company, was used as the oxygen carrier. To improve the mechanical strength, the oxygen carrier was calcined at 850°C under atmospheric air for 3 hr. The calcined oxygen carrier was mainly composed of 78 wt% Fe_2O_3 , 16.1 wt% SiO_2 , and 3.9 wt% Al_2O_3 , and no mercury was detected. Bituminous coal was selected as the solid fuel, and the proximate and ultimate analyses are shown in Table 1. The coal particles were sieved to a size range of 0.106–0.3 mm.

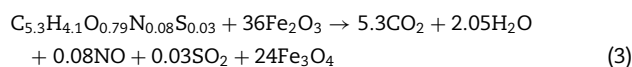
The amount of oxygen carrier used in experiments was determined by the balance calculation of oxygen carrier and fuel,

Table 1 – Proximate and ultimate analysis of coal.

Proximate analysis (wt%, ad)		Ultimate analysis (wt%, ad)		$\text{g } (\mu\text{g/g})$
M	1.46	C	63.49	1.122
V	14.36	H	4.11	
FC	45.87	O	12.79	
A	38.31	N	1.13	
		S	1.07	

M: moisture content; V: volatile matter; FC: fixed carbon; A: ash content; and ad: air-dried basis.

as shown in Eq. (3). Since 100 g coal contains 5.3 mol C, 4.1 mol H, 0.79 mol O, 0.08 mol N, and 0.03 mol S, the reaction between coal and oxygen carrier can be described as follows (Gu et al., 2015):



The ratio of coal to the Fe_2O_3 oxygen carrier was obtained according to the oxygen balance calculation. The calculation result indicates that when 100 g coal is fed into the FR, the weight of the oxygen carrier should not be less than 5760 g. In our experiments, the mass of coal was 0.5 g; hence, the theoretical amount of oxygen carrier was 30 g to ensure complete burning of coal.

1.2. Experimental setup

A schematic of the experimental facility is shown in Fig. 1, including a simulated gas generation system, a reaction system, and an analytical system. The simulated gas generation system consists of cylinder gases, mass flow controllers, and a vapor generator. The cylinder gases were provided by Nanjing Special Gas Co., Ltd. The gas flow rate was controlled by a mass flow controller (MFC, Beijing Horiba Metron Instruments Co., Ltd., China). The vapor generator was composed of an injection pump and a heating tap. The reaction system consisted of a quartz reactor, a heating furnace, and an intelligent temperature controller. The length and diameter of the reactor was 800 mm and 35 mm, respectively. A porous quartz plate was placed at the middle of the reactor to support the oxygen carrier and pulverized coal. The analytical system was composed of a filter, a condenser, a flue gas analyzer (ECOM J2KN, RBR Company, Germany), and a mercury analyzer (VM3000, Mercury Instrument Company, Germany). The exhaust gas of the reactor was first introduced to a filter for separating the particles, and then to a condenser for separating the vapors. Finally, the pretreated dry sample gas was measured using a gas analyzer and a mercury analyzer. All pipes and filters were made of Teflon to avoid mercury adsorption on the surface of the pipes. Heating tapes were also used before the reactor stage to preheat the gas and avoid vapor condensation. In addition, the mercury content in coal was directly analyzed with a mercury analyzer (Hydra II C, Leeman, USA).

1.3. Measurement

In order to ensure the accuracy of mercury measurement, before entering the mercury analyzer, the gas sample was first passed through NaOH solution to remove acidic gases (Cauch et al., 2008). For the measurement of Hg^0 , the gas sample was pretreated with KCl solution, in which Hg^{2+} could be absorbed. For the measurement of total gaseous mercury (Hg^{T}), the concentration of Hg^{2+} could not be directly obtained as the mercury content was determined online using a mercury analyzer via cold vapor atomic absorption spectroscopy, and SnCl_2 solution

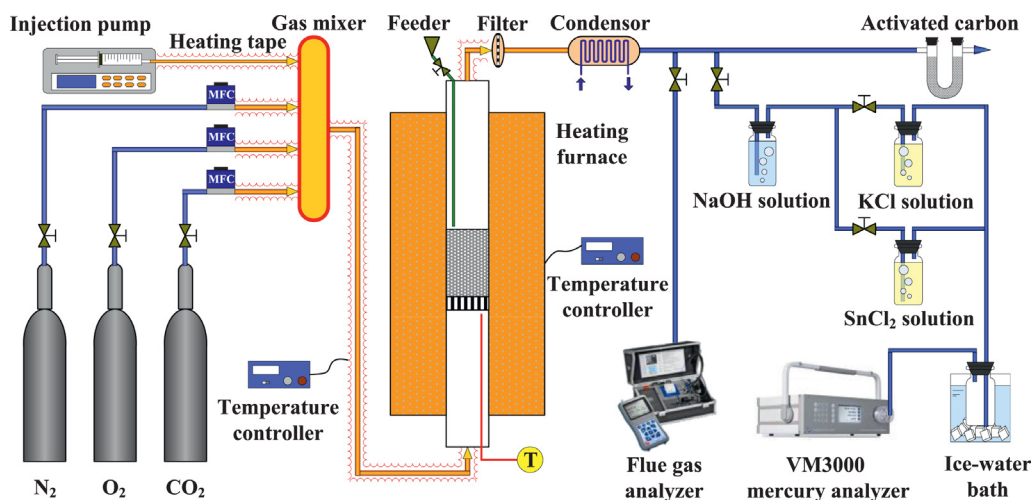


Fig. 1 – Schematic of experimental facility.

was used to pretreat the gas sample, in which Hg^{2+} could be oxidized to Hg^0 . For the measurement of Hg^{2+} , the concentration was obtained from the difference between the measured results of Hg^T and Hg^0 (Cauch et al., 2008).

1.4. Experimental procedures

In the experiment, 30 g oxygen carrier particles were placed on the porous plate and heated to the reaction temperature in N_2 atmosphere (2000 mL/min). Upon reaching the reaction temperature, the first cycle started in $\text{CO}_2/\text{H}_2\text{O}$ atmosphere (2000 mL/min). When the temperature re-stabilized, the coal was quickly introduced into the reactor zone. Thus, gaseous products generated from coal gasification reacted with the oxygen carrier, while the amount of oxygen carrier decreased. Meanwhile, mercury was gradually released from coal, which was monitored online using a mercury analyzer. The reaction duration was set to 10 min. After reduction, the oxygen carrier was oxidized in O_2/N_2 atmosphere (2000 mL/min) at the same temperature for 10 min. In the oxidation stage, the mercury release was again monitored online using a mercury analyzer.

2. Results and discussion

2.1. Effect of temperature on mercury release characteristics in FR

2.1.1. Effect of temperature on mercury release amount in FR
The reaction temperature is a critical factor affecting mercury release and speciation. It has been reported that gaseous mercury in flue gas mainly exists as Hg^0 and Hg^{2+} (Li et al., 2013b). Before the experiment, the chemical species of mercury in FR was predicted by a thermodynamic calculation via HSC Chemistry 6.0. The calculated results are shown in Fig. 2, which shows that the mercury in coal vaporized completely as soon as coal was added into the FR, and almost all mercury appeared as Hg^0 above 600°C. That is, owing to the high volatilization at a high temperature, the mercury in coal was easily transferred to gaseous products, which is similar to earlier research results (Frandsen et al., 1994). Fig. 3 provides the concentration of Hg released from FR. As shown in Fig. 3, with an increase in the reaction time, the concentrations of Hg^0 and Hg^{2+} released from the FR first increased, and then decreased slowly. A sharp peak of Hg^0 concentration can be observed after coal was added into the FR, the peak duration for which is approximately 180 sec. The results indicate that most mercury was released within 180

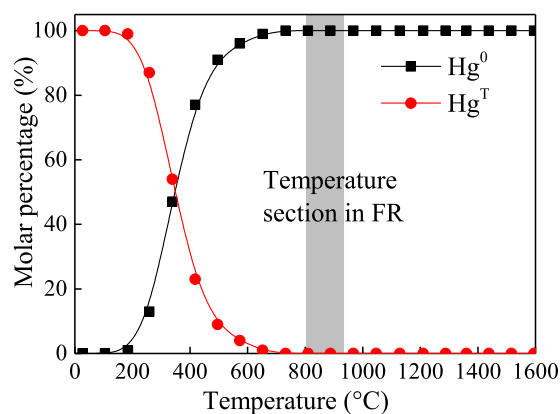


Fig. 2 – Thermodynamic calculation results of mercury chemical species at different temperatures in FR.

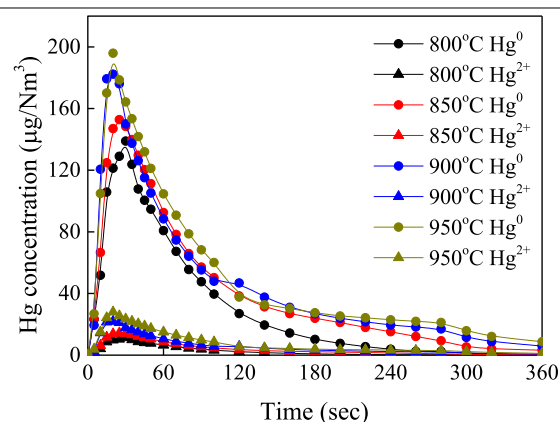


Fig. 3 – Concentration of Hg in flue gas as a function of reaction time in FR.

sec as Hg^0 in the FR at higher temperatures. In addition, Fig. 3 shows that the maximum concentration of Hg increased at a higher temperature in the FR.

Fig. 4 depicts the effect of reaction time on the amount of Hg^T released from FR. In our experiment, the temperature in FR ranged from 800–950°C when using CO_2 as the gasification medium. As observed, when the reaction time increased from 0

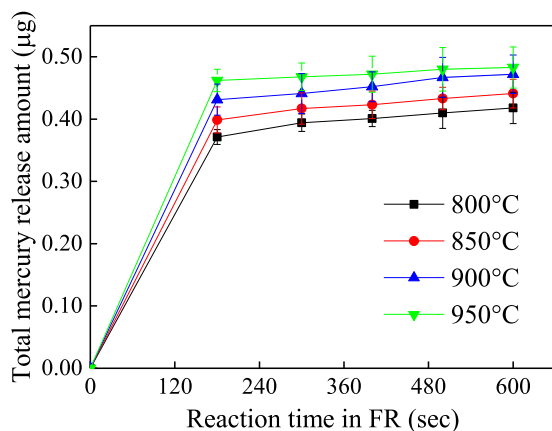


Fig. 4 – Effect of reaction time on mercury release amount in FR.

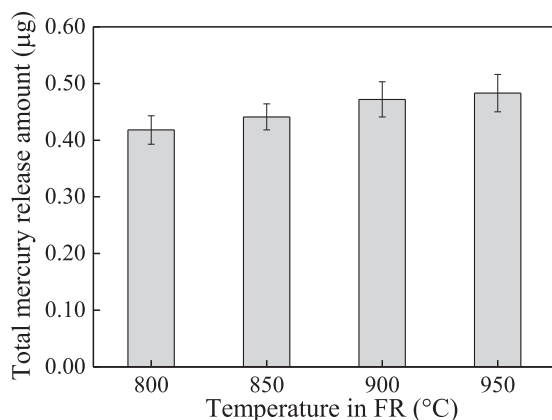


Fig. 5 – Effect of temperature on mercury release amount in FR.

to 180 sec, the released amount of Hg^{T} also increased. With an increase in reaction time, the released amount of Hg^{T} increased slowly, and then maintained a relatively stable value. The results indicate that when the reaction time reached 180 sec, most of mercury was released from coal. To ensure the complete release of mercury from coal, the reaction duration was set to 300 sec in all experiments.

Fig. 5 shows the effect of temperature on the amount of Hg^{T} released from FR. It should be noted that when the temperature increased from 800–950°C, the amount of Hg^{T} released from FR increased from 0.418 to 0.483 μg . However, the uptrend tended to slow down. As it has been proven, temperature is the decisive factor in mercury volatility (Li et al., 2011). Higher temperature is beneficial for increased release of mercury (Liu et al., 2010). Nevertheless, escaping channels might be reduced because of the decline in specific surface area of coal, which is caused by the solidification of coal at a higher temperature (Lin et al., 1994). In addition, the char would also act as activated carbon to absorb a part of the mercury from the gaseous products (Bai et al., 2010). That is, although more mercury is released from coal at a higher temperature, some amount might still be immobilized or reabsorbed by coal or char, resulting in a slight increase in released mercury with an increase in temperature.

2.1.2. Effect of temperature on mercury speciation in FR

Based on thermodynamic calculations, Hg^0 should be in a stable form in the high temperature region of FR. In FR, Hg^0 would remain as the dominant form. However, when the flue gas moves

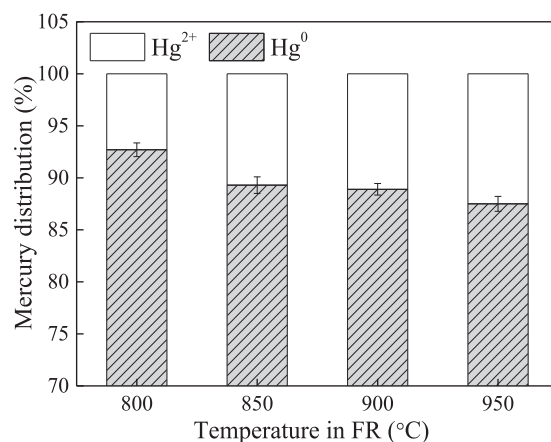


Fig. 6 – Effect of temperature on mercury distribution in FR.

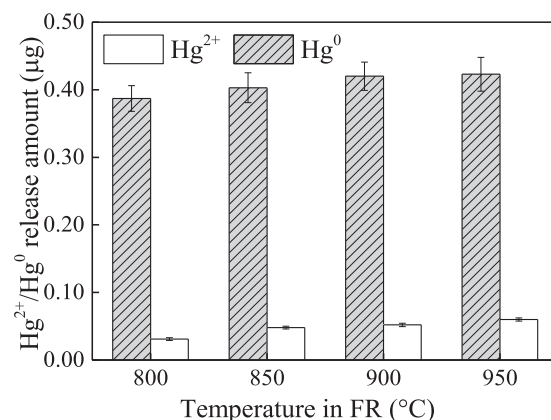


Fig. 7 – Effect of temperature on $\text{Hg}^{2+}/\text{Hg}^0$ release amount in FR.

out from the high temperature region, the temperature decreases and Hg^0 would react to form Hg^{2+} compounds. Thus, the conversion ratio of mercury was used in our experiment. Fig. 6 shows the mercury distribution in flue gas at different temperatures in FR. In our experiment, the temperature in FR ranged from 800–950°C, and CO_2 was used as the gasification medium. For each test, the reaction duration was set to 300 sec.

It can be observed from Fig. 6 that approximately 90% of mercury in the vapor phase is presented as Hg^0 in flue gas. It is believed that Hg^0 is a thermally stable form in a reducing atmosphere (Mendiara et al., 2014). Most of the mercury compounds will be converted to Hg^0 when the thermal decomposition temperature is higher than 700°C (Brown et al., 1999). However, Fig. 6 also shows that with an increase in the temperature in FR, although the mercury released from coal increased (see Fig. 7), the amount of Hg^0 decreased and the amount of Hg^{2+} increased. This can be attributed to different components of flue gas (Gao et al., 2013; Bao et al., 2008). It has been reported that the coal combustion flue gas in CLC process includes many components, such as SO_2 , CO , and Cl_2/HCl . According to earlier studies (Sliger et al., 2000; Zhou et al., 2007), the existence of Cl_2/HCl in flue gas is the main reason behind mercury migration and transformation. Hg^0 can react with Cl_2/HCl in flue gas to produce HgCl_2 as follows: First, Hg^0 reacts with Cl to form HgCl . Subsequently, HgCl is oxidized by Cl_2 into HgCl_2 , and generates Cl . When the temperature increases in FR, more Cl would be emitted from coal (Wang et al., 2017), causing more Hg^0 to be consumed by Cl_2/HCl , further leading to

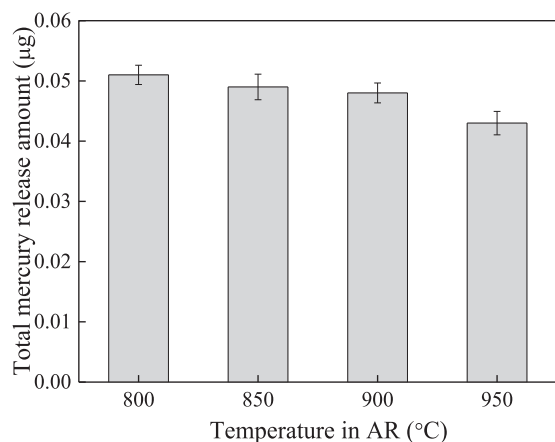


Fig. 8 – Effect of temperature on mercury release amount in AR.

an increase in released Hg^{2+} . Meanwhile, the CO concentration in flue gas would also decrease at a higher temperature (Wang et al., 2017). Since CO can inhibit the conversion of Hg^0 to Hg^{2+} (Lu et al., 2004), a low CO concentration implies that more Hg^0 could be oxidized to Hg^{2+} , leading to an increase in Hg^0 conversion rate. Thus, the proportion and amount of Hg^{2+} will increase when the temperature increases. Besides, oxygen carrier may also promote the conversion of Hg^0 to Hg^{2+} because of the existence of lattice oxygen (Ma et al., 2019). However, although a significant amount of Hg^0 is converted to Hg^{2+} in this process, it is still a stable form of gaseous mercury (Mendiara et al., 2014). Thus, only a part of Hg^0 would be oxidized to Hg^{2+} . As shown in Fig. 6, the conversion rate of Hg^0 to Hg^{2+} was about 10%.

2.2. Effect of temperature on mercury release characteristics in AR

2.2.1. Effect of temperature on mercury release amount in AR Fig. 8 illustrates the mercury release amount in the AR at different temperatures. In the AR, 5% O_2/N_2 was used as the agent and the temperature was consistent with that in the FR. As shown in Fig. 8, mercury was released in the AR, indicating that not all mercury in coal could be released in the FR. Approximately 90% of mercury in coal was released in the FR, while approximately 10% was released in the AR. This phenomenon can be explained as follows. 1) Coal did not burn completely in the FR, and some unburned coal or char entered the AR along with the oxygen carrier (Li et al., 2011). The unburned coal and char might have burned again in the AR because of the existence of O_2 , thereby releasing the remaining mercury (Ma et al., 2019). 2) The mercury released in the FR might have been adsorbed by the oxygen carrier. The porous structure and large surface area of the oxygen carrier, along with the produced Fe_3O_4 surface with reactive oxygen sites, was conducive to the physical and chemical adsorption of mercury (Guo et al., 2011; Li et al., 2016). Hence, when the oxygen carrier was oxidatively regenerated from molecular oxygen in the AR, the adsorbed mercury was desorbed.

Fig. 8 also shows that when the temperature increased from 800–950°C in AR, the released amount of mercury decreased from 0.051 μg to 0.043 μg . This was mainly because of the coal burning in FR. The coal burned more completely at a higher temperature in FR, resulting in lower amounts of unburned coal and char being circulated into AR (Mendiara et al., 2014). Consequently, the remaining amount of mercury in coal and char decreased. Thus, the amount of mercury released from the reburning of coal or char decreased in AR. Moreover, the higher temperature in FR also led to a reduction in the adsorption capacity

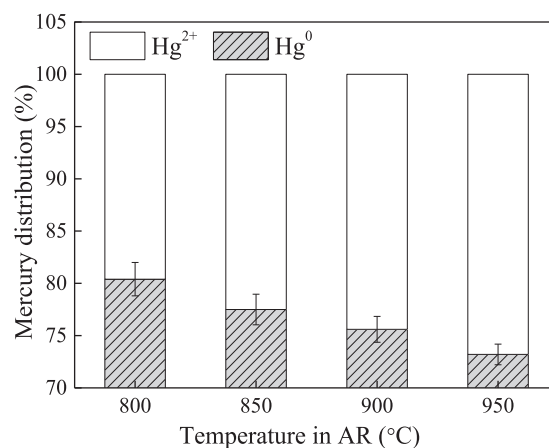


Fig. 9 – Effect of temperature on mercury distribution in AR.

of oxygen carrier for mercury. Since the mercury adsorbed by oxygen carrier in FR would be desorbed in AR, the amount of mercury released from oxygen carrier decreased.

2.2.2. Effect of temperature on mercury speciation in AR

Fig. 9 presents the mercury distribution at different temperatures in AR. It can be seen that the released mercury mainly existed as Hg^0 in AR, while Hg^{2+} only accounted for 20–25% of released mercury. That is, some mercury might have been oxidized in this process. In AR, 5% O_2/N_2 was introduced to regenerate the oxygen carrier. Simultaneously, the released mercury would also be oxidized by O_2 . Additionally, NO would also be generated in AR, which would also promote the oxidation of mercury at a low concentration (Yang et al., 2013). Although SO_2 could be produced in AR, the amount will be limited, which would minimize its inhibition effect on Hg^0 oxidation (Bao et al., 2008; Li et al., 2013a). Fig. 9 also shows that when the temperature increased in AR, the proportion of Hg^{2+} increased while the proportion of Hg^0 decreased. As reported before, the oxidation rate of Hg^0 is bound to temperature (Wang et al., 2016); thus, a higher temperature is beneficial for the conversion of Hg^0 to Hg^{2+} , causing an increase in the proportion of Hg^{2+} in AR.

2.3. Effect of gasification medium on mercury release in FR

In CLC process, the primary gas produced from FR contained H_2O (steam/vapor) and CO_2 , both of which are typical gasification mediums for coal burning. Hence, it is necessary to investigate the effect of gasification mediums on mercury release. In our experiments, the rate of $\text{H}_2\text{O}/\text{CO}_2$ ranged from 0:1 to 1:1 (g/L) and the temperature in FR ranged from 800–950°C.

Fig. 10 shows that the introduction of H_2O can improve the mercury release in FR. In addition, we also observe that the higher the ratio of $\text{H}_2\text{O}/\text{CO}_2$ in FR, the more the amount of released mercury. Compared with pure CO_2 as the reaction medium, the existence of H_2O would promote the emission of coal volatiles and reduce the melting of pore sizes on the coke particle surface, thus avoiding the reduction of mercury escape channels and improving the combustion efficiency of coal (Tian et al., 2014). Consequently, more mercury was released at a higher ratio of $\text{H}_2\text{O}/\text{CO}_2$ in FR.

However, Fig. 11 illustrates that the introduction of H_2O could improve the proportion of Hg^0 in released mercury. Correspondingly, it can also be inferred that a higher rate of $\text{H}_2\text{O}/\text{CO}_2$ would lead to a decrease in the proportion of Hg^{2+} . As reported earlier, the introduction of H_2O into FR would inhibit the formation of HgCl_2 , mainly because H_2O could inhibit the conversion of HCl to oxidative Cl in coal combustion, resulting in the inhibitory effect of mercury oxidation (Hu et al., 2017). That

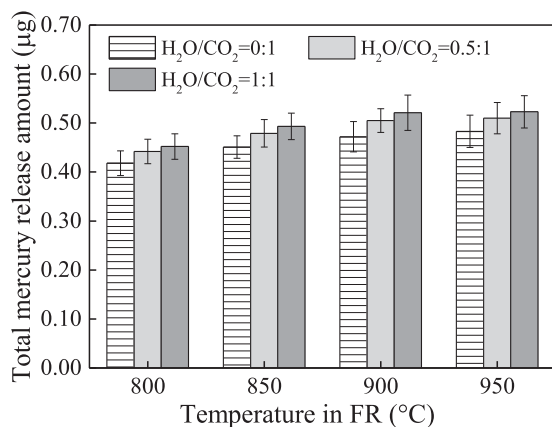


Fig. 10 – Effect of temperature on mercury release amount in FR at different H₂O/CO₂ rates.

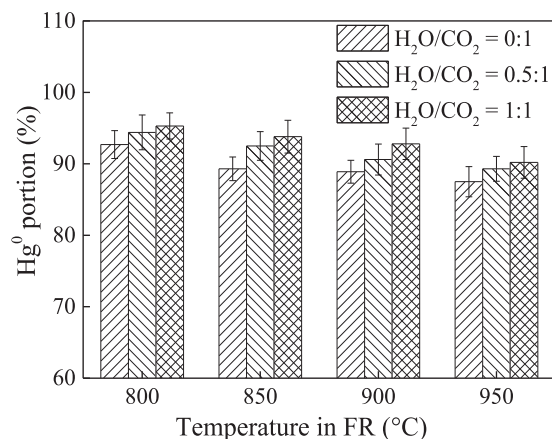


Fig. 11 – Mercury speciation in FR at different H₂O/CO₂ rates.

is, the path of mercury release in coal would change because of the introduction of H₂O (Sekine et al., 2008). First, Hg⁰ released from coal in FR could react with H₂O to produce Hg(OH)₂. Then, Hg(OH)₂ would easily change to HgO by heating. Finally, HgO would decompose into Hg and O₂ at a higher temperature. Based on these reasons, the proportion of Hg⁰ increased when the amount of H₂O increased in FR.

Meanwhile, Fig. 11 also illustrates that the proportion of Hg⁰ decreases when the temperature increases in FR. The results are in line with those in Fig. 6. This is mainly because of the different components of flue gas, as explained above.

2.4. Effect of reaction atmosphere on mercury release in the AR

In CLC, the reaction atmosphere mainly consists of O₂ and N₂. Usually, the O₂ concentration in the AR is less than that in air. Fig. 12 provides the mercury release amount in the AR at different O₂ concentrations. It can be seen that with an increase in the O₂ concentration from 5% to 21%, the mercury release amount almost remains constant, demonstrating that mercury in recirculated materials would be completely released in the AR (Mendiara et al., 2014), even with low O₂ concentrations. Fig. 12 also demonstrates that the total mercury release amount decreased with an increase in the temperature in the AR. These results are similar to those obtained in Fig. 8, and the reason has been explained above.

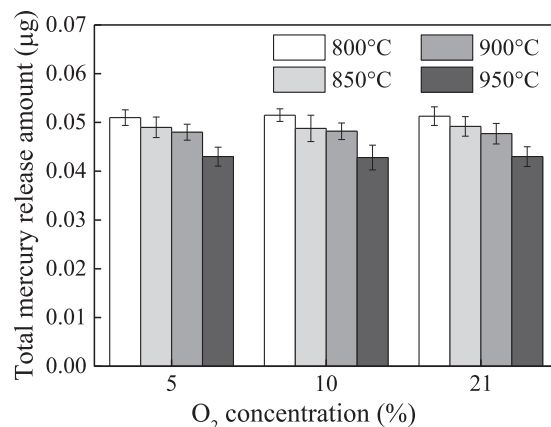


Fig. 12 – Mercury release amount in AR at different O₂ concentrations.

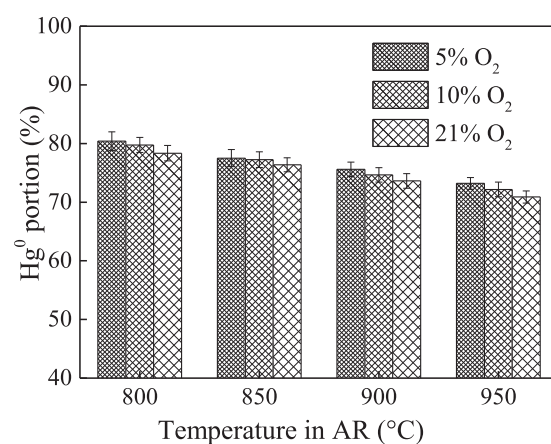


Fig. 13 – Mercury speciation in AR at different O₂ concentrations.

Fig. 13 depicts the mercury speciation in AR at different O₂ concentrations. The Hg⁰ proportion would decrease at a higher O₂ concentration. Correspondingly, the proportion of Hg²⁺ would increase. First, the increased O₂ concentration enhanced the oxidation of Hg⁰ (Li et al., 2017), thereby producing more Hg²⁺. Second, the higher O₂ concentration was advantageous for the complete burning of unburned coal or char, causing a decrease in the CO concentration in the AR. Hence, the inhibition effect of CO for Hg⁰ conversion was weakened (Ma et al., 2019; Lu et al., 2004), and more Hg⁰ could be converted to Hg²⁺. However, although more Hg⁰ could be converted to Hg²⁺ at a higher O₂ concentration, Hg⁰ could still exist as a stable form of gaseous mercury at higher temperatures. Consequently, the Hg⁰ proportion decreases with an increase in O₂ concentration, but the released amount of Hg⁰ is still higher than that of Hg²⁺. As shown in Fig. 13, the Hg⁰ proportion was always above 70% at different O₂ concentrations.

3. Conclusions

The knowledge of mercury release characteristics in CLC is critical for the development of this promising combustion technology. However, few studies have focused on this.

In this paper, the mercury release characteristics in the CLC process under different parameters such as FR temperature, AR temperature, FR gasification medium, and AR reaction at-

mosphere were investigated with bituminous coal, using Australian iron ore as the oxygen carrier in a fixed bed reactor. The results show that mercury could be released in CLC process. Approximately 90% of the mercury was released in FR, while about 10% was released in AR. Hg^0 was found to be the major form of released mercury from the CLC process. About 90% of mercury existed as Hg^0 in FR, and about 80% existed as Hg^0 in AR. A higher temperature in FR led to an increase in the total mercury released, and with an increase in temperature in FR, the proportion of Hg^0 decreased while that of Hg^{2+} increased. However, higher temperatures in AR resulted in a decrease in total mercury release amount and Hg^0 proportion. Gasification mediums in FR mainly included H_2O (steam) and CO_2 . The introduction of H_2O into FR was beneficial for the total release of mercury, but had an inhibiting effect on mercury oxidation. The total mercury release amount and Hg^0 proportion increased with an increase in the $\text{H}_2\text{O}/\text{CO}_2$ ratio of gasification mediums. The reaction atmosphere in AR mainly consisted of O_2 and N_2 . The higher O_2 concentration in AR had a negligible effect on total mercury release amount, but a positive effect on Hg^0 oxidation. With increased O_2 concentration, the total mercury release amount remained unchanged and Hg^0 oxidation was enhanced. Additionally, as a stable form at high temperatures, the released amount and proportion of Hg^0 were always higher than that of Hg^{2+} in CLC process under different conditions.

Considering the release of mercury in both AR and FR found in this study, the released amount of mercury in AR is much lower than that in FR, thus having a reduced effect on the environment than FR. Thus, further studies should pay more attention to reducing mercury release in FR.

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