

THE RELEASE OF MERCURY AND ARSENIC DURING THE COMBUSTION OF TWO CHINESE COAL GANGUES

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Abstract: The release ratios of mercury and arsenic during the combustion of two coal gangues with and without addition of CaO were studied at 600-1000°C. The experimental results showed that more mercury and arsenic released from two coal gangue samples with the increase in treatment temperature. At the same temperature, the release ratio of mercury was significantly higher than that of arsenic. The release ratio of arsenic decreased obviously after adding CaO, which indicates that CaO had a significantly inhibition effect on the release of arsenic. On the other hand, the CaO has a limited inhibition effect on the release of mercury during coal gangue combustion because of the release ratio of Hg slightly decreased after adding CaO.

Key words: Coal gangue, Arsenic, mercury, limestone, combustion

1. INTRODUCTION

Coal gangue is a kind of byproduct during coal mining or coal washing process. Generally, about 100-150 kg of coal gangue can be produced out of one ton coal (Liu & Liu, 2010), leading to huge amount of coal gangue piled near some coal mines in China, which cause serious environmental problems, such as occupying lands, polluting air, water, and soil (Querol et al., 2008; Zhou et al., 2010; Bell et al., 2000). Therefore, more and more attention has been paid on the re-utilization of coal gangue. For instance, some coal gangues with residue calorific value, has been utilized as solid fuel in some power plants in China, which was proven a reliable approach in reducing the quantity of coal gangue, so as to decrease its environmental hazards and make some economic benefits (Meng et al., 2013; Chugh & Patwardhan, 2004; Zhou et al., 2012). It was estimated that the utilization of coal gangue as primary fuel was up to 140 million tons in 2011 in China. However, the utilization of coal gangue in power plants has become a new anthropogenic emission source of mercury (Hg) and arsenic (As) due to the higher content of which in coal gangue (Zhou et al., 2015; Zhou et al., 2014).

Hg is considered as a toxic element that

seriously affects the ecosystem and human bodies (Swain et al., 2007; Sundseth et al., 2010; Pavlish et al., 2003; Lopez-Anton et al., 2010), which can cause bioaccumulation in the human body and thus be toxic to the central nervous system (Luo et al., 2009; Wang et al., 2010). As is also a toxic element because of its potential carcinogenic properties (Swaine et al., 2000; Guo et al., 2004). Due to its toxicity, the measures for controlling Hg and As emissions have been taken in coal-fired power plants since coal-fired power plants are one of the main anthropogenic sources of Hg and As emissions to the environment (Wang et al., 2010 and 2012). Among all the measures taken, the addition of CaO has been proved to be an effective way to control the Hg and As emissions during coal combustion (Jadhav & Fan, 2001; Sterling & Helble, 2003; Diaz-Somoano & Martinez-Tarazona, 2004; Li et al., 2007). However, the study concerning the addition of CaO for control Hg and As emission from coal gangue combustion is scarce.

In this study, in order to investigate the effect of CaO on the emission of Hg and As during coal gangue combustion, the emission of Hg and As from two Chinese coal gangues with and without addition of CaO were studied.

2. EXPERIMENTAL

2.1 Coal gangue samples

Two Chinese coal gangues (1 and 2) were used in this study, which were collected from Taiyuan second power plant and Taiyuan Coal washery in Shanxi province, respectively. The coal gangue samples were crushed, sieved to 0.16–0.27 mm, and dried prior to use. They were placed in plastic bags to prevent contamination and minimize oxidation. The proximate and ultimate analyses of the coal gangues and the concentration of Hg and As contents in the coal gangues are shown in Table 1.

2.2 Experiment procedures

The combustion experiments were conducted in a quartz boat located in a fixed bed tube reactor (20 mm inner diameter) under air atmosphere at temperature range of 600–1100°C. Approximately 2 grams coal gangue with the addition of CaO (Ca/S=1, 2 or 3) was charged into a quartz boat. Then it was quickly placed into the reactor at a predetermined temperature (600, 700, 800, 900, 1000 or 1100°C) with residence time of 40 min and an air gas flow of 300cm³/min. At last, the sample boat was moved to the cold end of the reactor immediately and cooled down in air flow. After the sample was cooled to room temperature, the combustion residue was weighed and used for Hg and As analysis.

2.3 Determination of Hg or As in coal gangues and combustion residue samples

The 0.2 g powdery coal gangue or combustion residue samples were precisely weighed and acid-digested with 10 mL oxidizing mixture (HNO₃:HCl = 3:1, volume ratio) in a Teflon digestion vessel. Then the digestion vessel was transferred into a microwave oven (MDS – 6G Microwave Apparatus) and was heated by the temperature programmed mode (130°C, 10 min; 150°C, 5 min; 180°C, 5 min; 200°C, 25 min) (Zhai et al., 2015); Next, the digested samples were diluted with double-distilled

deionized water; Then the Hg or As in sample is reduced with the potassium borohydride (KBH₄) and measured by AFS (Beijing Jinsuokun, SK-2003). The Hg analysis parameters of AFS is 380V of negative high voltage for photomultipliers, 30 mA of lamp Current, 600mL/min of Ar carrier flow, 800mL/min of Ar shield gas flow and 1% of KBH₄ concentration. The As analysis parameters of AFS is 280V of negative high voltage for photomultipliers, 80 mA of lamp Current, 600mL/min of Ar carrier flow, 800mL/min of Ar shield gas flow and 2% of KBH₄ concentration.

The release ratio (RR) of Hg or As was used to quantify the amount of Hg or As released from coal gangue during coal gangue combustion, which was calculated by the percentage of Hg or As released to the total Hg or As in coal gangue.

3. RESULT AND DISCUSSION

3.1 The release ratio of Hg or As from coal gangue at different temperature

3.1.1 The release ratio of Hg

Figure 1 shows the RR of Hg at different combustion temperature with and without addition of CaO (Ca/S=1) for two coal gangues. Figure 1 shows that the RR of Hg for two coal gangues without CaO has reached more than 95% at 600°C, which is similar to the law of the Hg release from coal combustion (Niu et al., 2012). It indicates that large amount of Hg in coal gangue can be released out below 600°C during coal gangue combustion. Also, it can be seen that the RR of Hg for both coal gangue samples slightly increases with temperature increasing. When the temperature increases to 1100°C, the RR of Hg reaches to about 96% and 99% for 1 and 2 coal gangue, respectively. Note that the increment of RR from 600°C to 1100°C is about 1% for 1 coal gangue and 3% for 2 coal gangue. This result illustrates that 2 coal gangue contains more thermal stable mercury compounds than 1 coal gangue, which possibly is caused by the fact that 2 coal gangue has more ash content than 1 coal gangue (Table 1).

Table 1. Proximate and ultimate analyses of the coal gangues

coal gangues	Hg in coal gangues ^b	As in coal gangues ^b	Proximate analysis, wt%				Ultimate analysis [daf], wt%			
			V _{ad}	A _{ad}	M _{ad}	C	H	N	S	O ^a
1	1.9	9	14.87	70.33	0.87	56.7	5.8	0.8	13.06	23.65
2	1.27	11.19	10.79	77.81	0.76	48.2	6.49	0.93	15.77	28.6

^a by difference. V: Volatile, A: Ash; M: Moisture; as: as received; daf: dry and ash free. ^b: ng/g

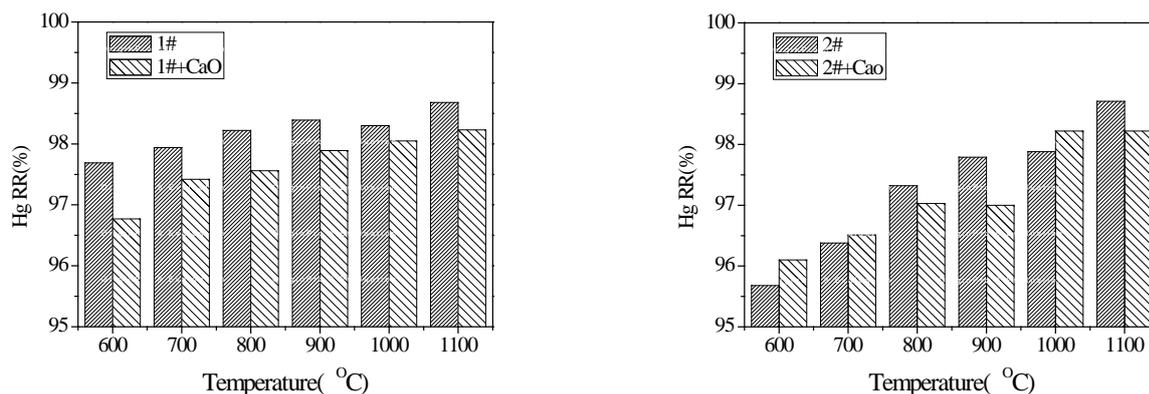


Figure 1. Release ratio of Hg during combustion of two coal gangues at different temperature

Figure 1 shows that the RR of Hg for two coal gangue with CaO was less than that without CaO under the same temperature. This agrees with the reports about the Hg release from coal combustion after adding CaO (Wang, 2006). It is reported that mercury can be removed by adding limestone in the circulating fluidized bed coal-fired units because adding excess CaO may generate strong chemical adsorption for HgCl_2 in flue gas (Niu et al., 2012). In addition, CaO has the potential to catalyze the reaction of mercury with chlorine in flue gas (Niu et al., 2012).

In this study, CaO can inhibit Hg release during coal gangue combustion, which should be ascribing to the similar reason. However, it should be noted that the decrement of RR with the addition of CaO is unobvious and it reaches to 95% at 600°C for two coal gangues. It indicates a limited inhibiting effect of CaO on mercury release from coal gangue, which possibly is attributed to the easily volatile character of mercury (Li et al., 2011; Roy et al., 2014).

3.1.2 The release ratio of As

The RR of As for two coal gangues is shown in Figure 2. It can be seen that the increase of temperature leads to an increase of As RR, which is in the range of 27.34%-42.1% and 19.83%-41.13% for 1

and 2 coal gangue, respectively. For 1 coal gangue, it shows a slight increase at 600-900°C with an increasing rate of 1.83%/100°C and a rapid increase at 900-1100°C that owns a 4.95%/100°C increasing rate. For 2 coal gangue, it increases slightly at 600-1000 °C with an increasing rate of 1.39%/100°C and increases rapidly at 1000-1100°C with an increasing rate of 15.76%/100°C. The rapid increase of As RR at higher temperature for two coal gangues indicates that high temperature can promote the As release. Also, it can be found that the As RR for 1 coal gangue is higher than that for 2 coal gangue at 600-1000°C, which indicates that the As compound in 1 coal gangue is less stable than that in 2 coal gangue. Overall, the As RR for the two coal gangues is similar, further suggesting that high temperature facilitates the release of As from coal gangue.

The RR of As for the two coal gangues with the addition of CaO is significantly lower than that without CaO at the same temperature range. Without CaO, it ranges from 27.34% to 42.1% for 1 coal gangue and from 19.83% to 41.13% for 2 coal gangue. 16.27% to 20.98% for 2 coal gangue after adding CaO. However, it decreases and ranges from 17.52% to 25.6% for 1 coal gangue and from this shows that the addition

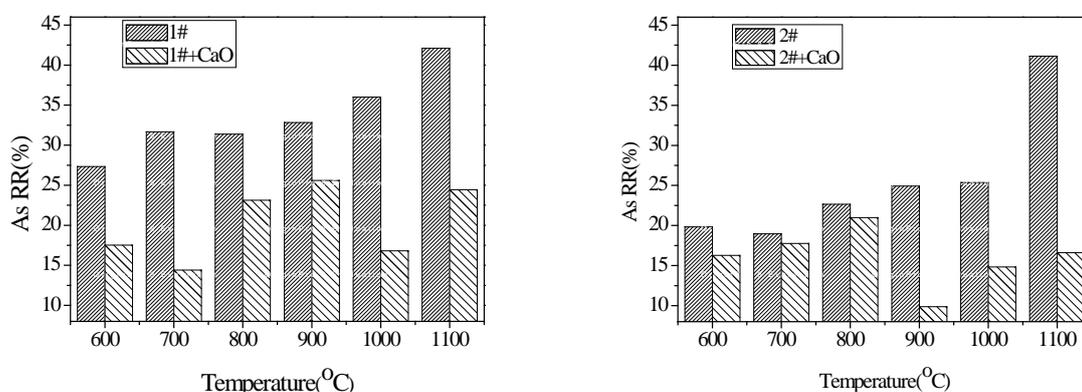
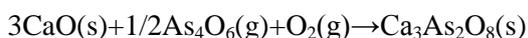


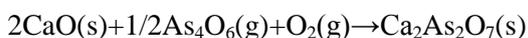
Figure 2. Release ratio of As during combustion of two coal gangues at different temperature

of CaO can significantly inhibit the release of As during coal gangue combustion, which is generally consistent with some reports about coal (Wang et al., 2003). According to the literature, there were some physical adsorption and chemical reactions between CaO and As during coal combustion (Diaz-Somoano & Martinez-Tarazona, 2004; Li et al., 2007). It was also reported that some chemical active sites on the surface of CaO played an important role on the inhibition of As from coal (Diaz-Somoano & Martinez-Tarazona, 2004; Li et al., 2007). Also, it was found that the addition of limestone inhibited the volatilization of As because the limestone decomposed into CaO at high temperature (Wang et al., 2003). Actually, the coal gangue is similar with coal because of their similar origin (Guo et al., 2012; Zhai et al., 2015; Zhang et al., 2015). Therefore, the decreased RR of As after the addition of CaO is possibly due to the similar reasons with that reported as coal, especially the active sites on the surface of CaO as reported about coal (Wang et al., 2003). According to the report about coal, the active sites on the CaO possibly had some reactions with As element, such as the following chemical reactions, leading to the decrease of As release from the two coal gangues (Diaz-Somoano & Martinez-Tarazona, 2004; Li et al., 2007):

Below 700°C



At 800°C~1100°C



If greater than 1100°C, $\text{Ca}_2\text{As}_2\text{O}_7$ will be converted into $\text{Ca}_3\text{As}_2\text{O}_8$.



Actually, the products of the reactions, such as $\text{Ca}_3\text{As}_2\text{O}_8$ are thermal stable and cannot decompose at the temperature range studied (Diaz-Somoano & Martinez-Tarazona, 2004). In this way, the As release from coal gangue can be inhibited by the addition of CaO.

3.1.3 The comparison of Hg RR and As RR

According to Figure 1 and Figure 2, it can be found that temperature is one of the key factors influencing the release of As and Hg. This is consistent with the result reported in the literatures (Bai, 2003; Liu et al., 2000; Gao et al., 2016; Wang et al., 2002; Yokoyama et al., 2000). Generally, with the increase of heating temperature, the volatile matter from coal gangue also released rapidly, which enlarged the porosity in the coal gangue and accelerated the diffusion of the gaseous Hg and As, thus resulting in the increased release of Hg and As. However, it should be noted that the RR of Hg is significantly higher than that of As at the same heating condition. It indicates that Hg is more volatile than As during coal gangue combustion, which agrees with the result during coal combustion (Zhou et al., 2012). Actually, Hg is an extremely volatile element and most of Hg in coal or coal gangue will release to the gas phase during coal or coal gangue combustion (Lv et al., 2004; Chu & Porcella, 1995). Compared with Hg, As is a relatively stable element (Zhou et al., 2012). Therefore, it can be observed a higher RR of Hg than that of As. Meanwhile, the addition of CaO can effectively inhibit the release of As while it only slightly inhibit the release of Hg, which is also agreement with the Hg and As release behavior during coal combustion (Zhai et al., 2015; Li et al., 2011). It further suggests the easily volatile character of Hg no matter it is in coal or in coal gangue (Zhai et al., 2015; Li et al., 2011).

To clearly show the result, the inhibition rate was used to express the inhibition effect of Hg and As release after addition of CaO, which was defined as the difference between the RR of Hg or As with and without the addition of CaO. And the result is shown in table 2 and table 3.

Table 2 shows that the inhibition rates of Hg are all less than 1%, indicating an inconspicuous inhibition effect of CaO on the release of Hg. As we all know, Hg is a kind of volatile element and can easily release out during coal gangue combustion (Zhai et al., 2015). Therefore, the addition of CaO has no distinct inhibition effect on Hg release from coal gangue.

Table 2. The inhibition rate of Hg at different temperature

coal gangue	600°C	700°C	800°C	900°C	1000°C	1100°C
1	0.92	0.52	0.66	0.5	0.25	0.45
2	-0.42	-0.13	0.29	0.79	-0.3	0.49

Table 3. The inhibition rate of As at different temperature

coal gangue	600°C	700°C	800°C	900°C	1000°C	1100°C
1	9.82	17.26	8.27	7.22	19.2	17.7
2	3.56	1.21	1.68	15.05	10.5	24.5

However, table 3 shows a significantly inhibition effect of CaO on the release of As. Meanwhile, the inhibition rate of As shows a strong dependency on the temperature. For example, the inhibition effect for 2 coal gangue is not obvious below 800°C and its highest inhibition rate is only 3.56%. However, at temperature >900°C, the inhibition effect significantly enhanced and the minimum inhibition rate reached to 10.54%. This further reflects that some reactions between As and CaO mentioned above can be occurred and some more stable As compound such as $\text{Ca}_3\text{As}_2\text{O}_8$ can be produced at higher temperature for 2 coal gangue (Diaz-Somoano & Martinez-Tarazona, 2004). For 1 coal gangue, the inhibition effect at 600 °C, 800 °C and 900°C is similar with the lowest inhibition rate of 7.22% while the inhibition effect at 700°C, 1000°C and 1100°C is similar with the highest inhibition rate of 19.2%. It indicates that the inhibition effect of CaO on the release of As is different with different type of coal gangue, which is possibly caused by the different modes of occurrence of As existed in the two coal gangues (Cao et al., 2017).

3.2 The RR of Hg and As from two coal gangues with CaO addition in different Ca/S ratio

In order to further study the effect of CaO on the inhibition effect of Hg and As release, the CaO addition in different Ca/S ratio (Ca/S=1, 2 or 3) was adopted during coal gangue combustion at 800 °C, 900 °C and 1000°C, respectively.

3.2.1 The RR of Hg with CaO addition in different Ca/S ratio

The RR of Hg with CaO addition in different Ca/S ratio is shown in Figure 3. It can be seen that there is no obvious change for the RR of Hg at different temperature with different Ca/S ratio for the two coal gangues. As stated previously, the RR

of Hg can reach to more than 95% at 600°C or higher temperature and the inhibition effect of CaO on the mercury in coal gangue is not obvious. The results of RR of Hg with CaO addition in different Ca/S ratio confirm the above conclusions. This further indicates that CaO has a limited effect on the Hg release no matter how much CaO was added. Also, it implies that the increase of the CaO amount has nearly no distinct effect on the controlling Hg release from coal gangue.

3.2.2 The RR of As with CaO addition in different Ca/S ratio

As shown in Figure 4, CaO has different effect on the inhibition of As release with CaO addition in different Ca/S ratio. In most cases, it can significantly decrease the As release with the increase of CaO addition, i.e. the more CaO used, the more As inhibited. However, the difference of Ca/S ratio presents different effect on As release for the two coal gangues. For example, the RR of As in Ca/S of 1 is higher than that in Ca/S of 3 and lower than that in Ca/S of 2 at 800°C for 1 coal gangue. Meanwhile, the RR of As in Ca/S of 3 is lower than that in Ca/S of 2 and higher than that in Ca/S of 1 at 900°C for 2 coal gangue. Overall, the suitable Ca/S ratio at each temperature for As inhibition is different for each coal gangue. For example, the suitable Ca/S ratio for As inhibition were Ca/S of 3 at 800°C, Ca/S of 1 at 900°C and Ca/S of 2 at 1000°C for 1 coal gangue. For 2 coal gangue, the suitable Ca/S ratio for As inhibition is Ca/S of 3 at 800°C, 900 °C and 1000 °C. It is probably caused by the different modes of occurrence of As with different thermal stable stability existed in two coal gangues (Liu et al., 2009). In summary, the performance of CaO on the As inhibition is mainly affected by the ratio of Ca/S and temperature. Also, it has relation with the mode of occurrence of As in coal gangue (Cao et al, 2017).

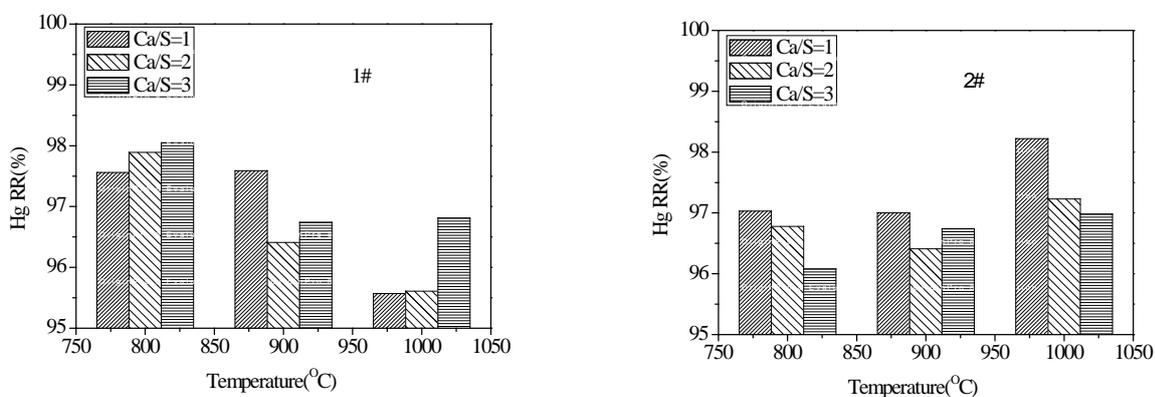


Figure 3. RR of Hg during combustion of two coal gangues with CaO addition in different Ca/S ratio

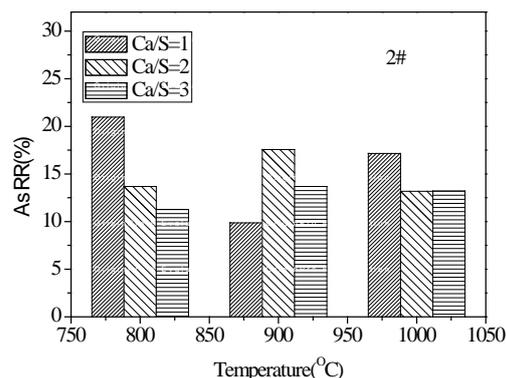
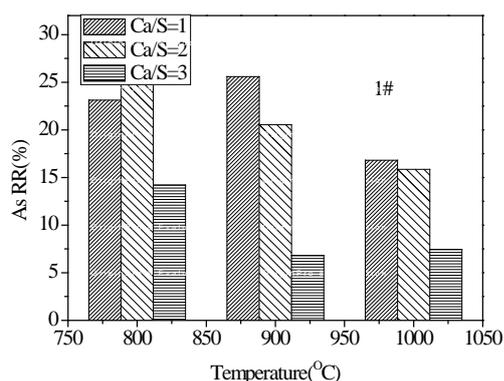


Figure 4. RR of As during combustion of two coal gangues with CaO addition in different Ca/S ratio

4. CONCLUSION

The release ratios of Hg and As during two coal gangues combustion were studied and the following conclusions can be drawn:

1. For the two coal gangues, the release of Hg and As were gradually increasing with the increase of temperature. The Hg RR is significantly higher than the corresponding As RR, indicating that the volatility of Hg in coal gangue is much higher than that of As.

2. According to the difference of Hg RR and As RR with and without addition of CaO, it can be found that the addition of CaO has some inhibition effect on the release of Hg. However, the inhibiting effect of CaO on Hg release from coal gangue is limited because of the easily volatile character of Hg. The inhibition effect of CaO on the release of As is much more than that on the release of Hg because some reactions between As and CaO can be occurred and produce some more stable As compound.

3. The results with CaO addition in different Ca/S ratio indicates that CaO has a limited effect on the Hg release no matter how much CaO was added for the two coal gangues. And the As inhibition is mainly affected by the ratio of Ca/S and temperature as well as the mode of occurrence of As in coal gangue.

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