Effect of oxidation treatment on the adsorption and the stability of mercury on activated carbon

HU Chang-xing, ZHOU Jin-song*, LUO Zhong-yang, HE Sheng, WANG Guang-kai, CEN Ke-fa

(State Key Laboratory of Clean Energy Utilization, Zhejiang University, Hangzhou 310027, China. E-mail: zhoujs@cmee.zju.edu.cn; huchx@zju.edu.cn)

Abstract: Oxidation treatment on the adsorption and the stability of Hg on activated carbon (AC) was investigated. Both MnO_2-AC and $FeCl_3$-AC were produced during oxidation treatment. The measurement of modified AC's mercury adsorption capacity was conducted in a simulated coal-fired flue gas by adsorbing test apparatus. TCLP and column leaching methods were used to test the stability of mercury adsorbed on ACs. The results indicate that the oxidation treatment changed the pore structure of the AC and modified the carbon surface by creating chemical components such as MnO_4^-, Mn^{4+}, O, NO_3^-, Fe^{3+}, Cl^-, etc. The Hg sorption capacity on MnO_2-AC or $FeCl_3$-AC was about three times higher than that of untreated carbon. In addition, the mercury control cost of each of the formers was about the half cost of the untreated carbon. The stability of Hg absorption was studied, it found that mercury adsorbed on the oxidation treated AC was not better than that of untreated carbon. It could concluded that the insoluble form of Hg is very important to the stability of mercury adsorbed on AC. This study suggests that the $FeCl_3$-AC is the best absorbent for Hg with high adsorption capacity, better Hg adsorption stability in leaching environment, and lower cost among the three ACs tested.

Keywords: activated carbon; oxidation treatment; Hg adsorption; stability

Introduction

Mercury (Hg) pollution becomes a major global environmental concern lately due to the highly toxic pollutant is wide distributed globally through long-range transport. The environmentally persistent, bio-accumulative mercury has a strong, long-term human health impact, for children in particular, resulting from food chain exposure. Furthermore, China is the world's biggest coal producing and consumption state, both the environmental impact and reduction of Hg from coal combustion are particularly important for China.

Coal-fired utility boilers are currently one of the largest sources of Hg emissions and Hg emission controls are required during coal combustion. Injection of activated carbon (AC) upstream of either an electrostatic precipitator (ESP) or a fabric filter (FF) is a retrofit technology that has been in the phase of field testing for controlling coal-fired power plant Hg emissions in the USA (Pavlish et al., 2003). However, the projected cost of Hg control by AC injection is significant (USEPA, 1997; Brown et al., 1999; Pavlish and Mann, 1998). The costs for injection of AC for achieving a Hg reduction rate of 90% varies approximately from 0.03 to 0.4 cents/kWh, according to estimates made by Environmental Protection Agency (USEPA, 2000; Kilgroe and Srivastava, 2000). Applying a modified activated carbon with higher Hg adsorption capacity may bring this cost down greatly. In addition, the mercury's stability on adsorbent is critical for avoiding Hg secondary pollution when using the AC injection

Hg control technology.

It is well known that there are three forms of Hg existed in coal combustion flue gas: elemental mercury (Hg^0), oxidized forms of mercury (Hg^{2+}), and particle-bond mercury (Hg_p) (USEPA, 2002). The water soluble Hg^{2+} is easily captured by wet scrubbers, and Hg_p can be collected by particle control devices. However, Hg^0 is very difficult to capture for its high volatility and insolubility in water. The purpose of this research is to study the effect of improved oxidation treatment method on the Hg adsorption capacity of AC and the stability of Hg adsorbed on the modified AC.

1 Experimental

1.1 Preparation of AC by oxidation treatment

A commercially available AC, produed from wood by the zinc chloride method (Shanghai Activated Carbon Co., Ltd., 2004) was used. The AC was dried in an electric oven at 363 K, cooled down to room temperature, and then stored in a desiccator for use, which was named BLC.

The BLC was treated by using two different oxidizing solutions: an active MnO_2 solution and a $FeCl_3$ solution. During BLC treatment by the active MnO_2 solution, 20 ml concentrated nitric acid (HNO_3) was put into 200 ml deionized water. After that, 5 ml 50% manganous nitrate solution (50% $Mn(NO_3)_2$) was mixed before 2.76 g of potassium permanganate ($KMnO_4$) was added. Then the active MnO_2 solution was prepared. Finally, 100 g of BLC was mixed with the active MnO_2 solution, continuously stirred about 30 min, dried in an electric oven at a temperature of

363 K, cooled down to room temperature, and then stored in a desiccator for future use, which was named MnO_2-AC. Similarly, the $FeCl_3$ solution was prepared by mixing 20 ml of concentrated hydrochloric acid (HCl) with 200 ml of deionized water, and then, adding 5 g ferric trichloride ($FeCl_3$) solid reagent. At last, 100 g BLC was mixed with the $FeCl_3$ solution, stirred, dried, cooled and stored as the above. In that way, $FeCl_3$-AC was ready.

1.2 Mercury adsorption

The experiment of measuring AC's Hg adsorption capacity and the one for preparing a Hg-laden AC were conducted in a simulated coal-fired flue gas using adsorbing apparatus. The schematic diagram of the experimental system is shown in Fig.1. A simulated flue gas typically containing 6% oxygen (O_2), 12% carbon dioxide (CO_2), 20 ppm nitrogen dioxide (NO_2), 300 ppm nitric oxide (NO), 1500 ppm sulfur dioxide (SO_2), and 50 mg/L hydrogen chloride (HCl), was used in this research, with nitrogen (N_2) for balance and carrying Hg^0. The Hg^0 in the simulated flue gas was generated by means of a mercury permeation tube (made by VICI Metronics Inc, USA). The concentration of Hg^0 was controlled by altering the temperature of the permeation tube and the flow rate of the Hg^0 carrying gas.
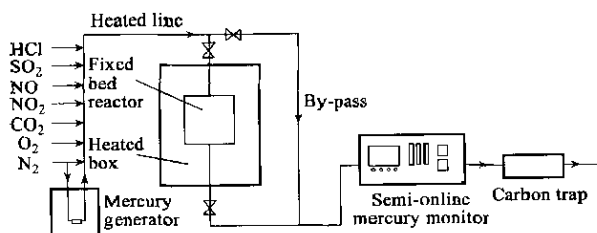

Fig.1 Schematic diagram of the adsorption experimental system

Before measuring AC's Hg adsorption capacity was started, a 0.002-g AC with 1 g powdered quartz, which can help AC equally distribute, was placed in the fixed-bed reactor and the temperatures of the heated line and the heated box were maintained at 393 K. Then the flow of simulated flue gas was adjusted to 1 min^{-1} with the typical composition stated above. The Hg^0 in the simulated flue gas was controlled at a desired concentration, which was about 65 $\mu g/Nm^3$. During the experiment, a semi-online mercury monitor QM 201 automatically recorded the concentration of Hg^0 in the simulated flue gas every 9 min.

According to the experience with measuring AC's Hg adsorption capacity, a 2-g AC was used during the experiment for preparing a Hg-laden AC. the Hg^0 concentration was maintained at about 500 $\mu g/Nm^3$. The experiment time, which was generally controlled at about 208 min, was adjusted and determined by the Hg^0 concentration in the exhaust gas in order for the

amount of Hg adsorbed by AC to reach (52 ± 1) $\mu g/g$. The other experimental conditions were the same as those of measuring AC's Hg adsorption capacity. When each preparing experiment was finished, 1 mg Hg-laden AC was picked to examine the Hg concentration. If the Hg concentration on AC did not accord with the (52 ± 1) $\mu g/g$, the Hg-laden AC would be not used in the Hg stability experiment.

1.3 Leaching methods

A variety of laboratory leaching methods for Hg have been developed, including batch tests, column tests, and serial and sequential leaching tests, etc. In this study, two different leaching methods, batch test and column test, were used. The batch test was carried out using a standard leaching protocol called the toxicity characteristic leaching procedure (TCLP), which is an accelerated experimental method designed to simulate conditions in an anaerobic landfill and involves leaching a mass of waste material by an acetic acid solution, using a 20:1 liquid-to-solid mass ratio, and rotating the mixture for (18 ± 2) h at 30 r/min (Luo *et al.*, 2006). The details of TCLP are available elsewhere (USEPA method 1311, 1992). About 5 g of AC was used in each test. Five different leaching liquids were used in the column tests to simulate various disposal solution environments. They included deionized water, 1% H_2SO_4, 1% $FeCl_3$, 1% Na_2CO_3, and 1% NaOH. The flow of leaching liquid was controlled at about 5 ml/h and the leachate was collected everyday for analysis during the column tests.

1.4 Instrument analysis

The surface characteristics of ACs were mainly determined by using an Autosorb-1-C, which is a kind of automated chemisorption/physisorption analyzer for surface area and pore size measurements, supplied by Quantachrome Instruments, USA. The surface chemical compositions of ACs were analyzed by using a JEOL JEM-2010 electron microscope with the INCA energy dispersive X-ray microanalysis. Microwave digestion was used for complete dissolution of ACs. The mercury concentration in the digestion solution was determined by using a XGY-1011A atomic fluorescence spectrometer, made by the Institute of Geophysical and Geochemical Exploration under China Academy of Geosciences. It is based on cold vapor atomic fluorescence spectroscopy (CVAFs) and its detection limit <0.01 $\mu g/L$ with relative standard deviation (RSD) <1.8%. The QM 201 instrument was used to measure the gaseous Hg^0. The principle of QM 201 operation is the same as that of XGY-1011A. Its measuring range is 0.01—100 $\mu g/L$, detection limit is not bigger than 0.003 $\mu g/L$, and reproducibility is lower than 5%.

2 Results and discussion

2.1 Effect of oxidation treatment on AC

The multipoint Brunauer-Emmett-Teller (BET) surface areas of the carbons were measured by N_2 adsorption at 77 K with relative pressures (P/P_0) up to 0.99, using Autosorb-1-C automated analyzer. The total pore volumes of the samples were evaluated according to the N_2 adsorption isotherms at $P/P_0=0.99$, and the micropore volumes were estimated according to the N_2 adsorption isotherms at 77 K using the Dubinin Astakhov (DA) Method.

The results of surface characteristics of the three ACs (BLC, MnO_2-AC, and $FeCl_3$-AC) are shown in Table 1. The specific surface area and the pore volume of both modified ACs, especially MnO_2-AC, were less than those of BLC. The bulk specific weight was increased for both MnO_2-AC and $FeCl_3$-AC. It means

that the surface characteristics of BLC were changed as a result of impregnated oxidation treatment. However, there are clear differences in surface characteristics between MnO_2-AC and $FeCl_3$-AC as a result of different impregnated oxidization approaches. The use of $Mn(NO_3)_2$ and $KMnO_4$ in a concentrated HNO_3 solution during the process of preparation of MnO_2-AC may have both effects of oxidizing the carbon as well as impregnated the carbon with the manganese compounds, which results in significant reductions in porosity and surface area. On the other hand, $FeCl_3$ is not a strong oxidizing agent which does not oxidize the AC during $FeCl_3$/HCl treatment process. Thereby, the porosity and surface area of $FeCl_3$-AC just slightly changed owing to the impregnated agents remained on the internal surface

Table 1 Surface characteristics of activated carbons

Activated carbon	Specific surface area, m^2/g	Bulk specific weight, g/cm^3	Total pore volume, cm^3/g	Micropore volume (<2 nm), cm^3/g	Particle size, mm	Porosity, %
BLC	1850	0.35	1.05	0.35	0.09	67
MnO_2-AC	865	0.41	0.29	0.10	0.09	45
$FeCl_3$-AC	1470	0.38	0.92	0.31	0.09	58

after the treatment. To better understand the significant difference in surface characteristics between MnO_2-AC and $FeCl_3$-AC, further research is needed.

Fig.2 shows the main elements detected on the surface of ACs (Cu was not detected on the surface of ACs, but was presented in the AC holder during analysis). Table 2 shows the chemical composition on

the surface. The results show no Mn and Fe existed on the surface of BLC. More O and Mn presented on the surface of MnO_2-AC than that on the surface of BLC as a result of Mn $(NO_3)_2$/$KMnO_4$/HNO_3 treatment. Meanwhile, the $FeCl_3$-AC showed significant increases in Fe and Cl contents with very little change in O content of the treated carbon due to the impregnated $FeCl_3$. All these results confirm that the
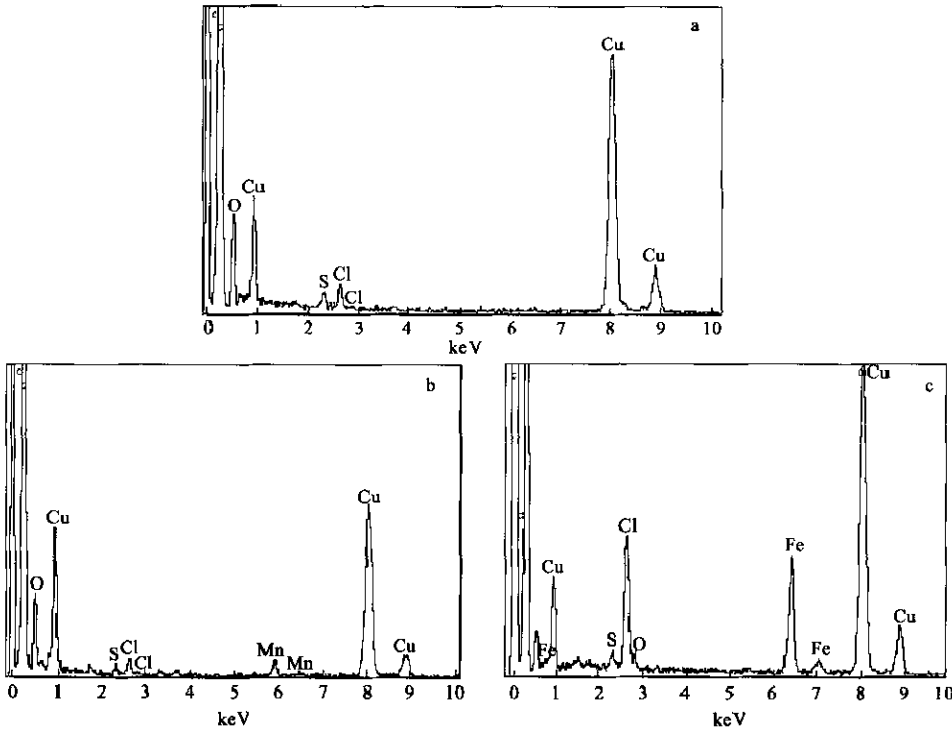

Fig.2 Main elements detected on the surface of ACs BLC (a), MnO_2-AC (b) and $FeCl_3$-AC (c)

oxidation treatment by an active MnO_2 solution or a $FeCl_3$ solution on the surface of BLC not only changes the surface physical structure of the BLC, but also modifies the surface chemical composition of the carbon. However, it is not clear that the increases in O and Mn are due to oxidation and/or impregnation of chemical agents which capable both oxidize and impregnate the carbon. The chemical forms of these elements existed on the surface are not known. It is speculated based on the original oxidant solution used for the treatments that these functional groups may exist as MnO_4^-, Mn^{4+}, O, NO_3^-, Fe^{3+}, Cl^-, or other types. Further research is needed in order to understand more about the exact forms of these groups on the surface of ACs.

Table 2 Surface chemical composition of activated carbon (wt%)

Element	C	O	S	Cl	Mn	Fe	Others
BLC	93.96	4.4	0.55	0.57	0	0	0.52
MnO_2-AC	80.48	12.81	0.43	0.4	4.64	0	1.24
$FeCl_3$-AC	84.44	4.39	0.44	4.67	0	5.57	0.49

2.2 Hg sorption capacity of ACs

The bench-scale, fixed-bed test was performed to evaluate the mercury sorption capacity of BLC, MnO_2-AC and $FeCl_3$-AC according to the description of Section 2.1. Fig.3 presents the results of the Hg sorption capacity for BLC, MnO_2-AC and $FeCl_3$-AC. In the first 20 min, the curvilinear trends of three ACs were very similar. After that, it only took BLC about 50 min to reach the Hg saturation adsorption, which was about 1100 μg/g. However, the trend of MnO_2-AC Hg sorption capacity curve was similar to that of $FeCl_3$-AC curve, which took about 160 min to reach Hg saturation adsorption of 3300—3400 μg/g. The results imply that the functions of Mn $(NO_3)_2$/ $KMnO_4$/HNO_3 treatment and $FeCl_3$/HCl treatment are similar in the way of enhancing the Hg sorption capacity of AC.

From Fig.3, it can be seen that the Hg sorption capacities of MnO_2-AC and $FeCl_3$-AC, are about three times higher than that of the BLC. Previous researches (Ren, 2003; Karatza *et al.*, 2000) found that ACs with higher surface area and more micropore would have higher adsorption capacity. However, the results of

Fig.3 Hg^0 sorption capacity of ACs

this study (Fig.3 and Table 1) are not in accord with the previous researches. The specific surface area, the total pore volume, and the micropore volume of the MnO_2-AC were all less than those of BLC, whereas the Hg sorption capacity of MnO_2-AC was much higher than that of BLC. In addition, the surface characteristic of $FeCl_3$-AC was similar to that of BLC, but BLC's sorption capacity of Hg was much less than that of $FeCl_3$-AC. Based on the data shown in Fig.2 and Table 2, the surface chemical characteristics of the three ACs tested were different. It can be assumed that the surface physical characteristic of AC is just one of the factors that determine Hg adsorption. The possible oxidative groups, MnO_4^-, Mn^{4+}, O, Fe^{3+}, and others, remained on the surface after impregnated oxidation treatment may oxidize Hg during the Hg adsorption process and resulting in the higher Hg sorption capacity of the MnO_2-AC or $FeCl_3$-AC.

2.3 Stability of Hg on ACs

Injection of AC upstream of either an ESP or FF is regarded as one of the feasible retrofit Hg control technologies in the near future. This method controls Hg emissions from coal-fired power plants by adsorbing Hg in the flue gas by the injected AC. Hence, the stability of Hg adsorpted by AC is very important for avoiding Hg release after the adsorption.

Fig.4 shows the leaching results of BLC, MnO_2-AC, and $FeCl_3$-AC with five different leaching liquids in continuous 14 d. Results for the standard TCLP test are also included. Fig.4a shows the average Hg concentration in 1% NaOH leachate (pH>14) was about 0.89 μg/L, which was close to the average Hg concentration (about 0.78 μg/L) in 1% H_2SO_4 leachate (pH<1); however, this value was much higher than the average Hg concentrations in H_2O, 1% $FeCl_3$, and 1% Na_2CO_3 leachates (3<pH<10), which mostly ranged from 0.22 to 0.28 μg/L. It seemed that Hg was easily released from the surface of the BLC in the strong alkaline or acid solution environment. Compared with the Hg concentration of the TCLP leachate, about 0.73 μg/L, the Hg concentrations in all the leachates were still lower than the safe limit defined by the Universal Treatment Standard (accessed June, 2004), which limits content of mercury in the TCLP leachate to 25 μg/L. Based on these results, a conclusion can be drawn that the Hg is stable on the surface of the BLC in the solution environment.

The leaching results of MnO_2-AC are shown in Fig.4b. The average Hg concentrations in the leachates of H_2O, 1% H_2SO_4, and TCLP are significantly higher than the safe limit of 25 μg/L. In addition, although the Hg concentrations in the leachates of 1% $FeCl_3$, 1% Na_2CO_3, and 1% NaOH are much lower than those in the leachates of H_2O, 1% H_2SO_4, and TCLP, however, they are still much higher than those for the BLC in Fig.4a. This indicates that the Hg adsorbed or
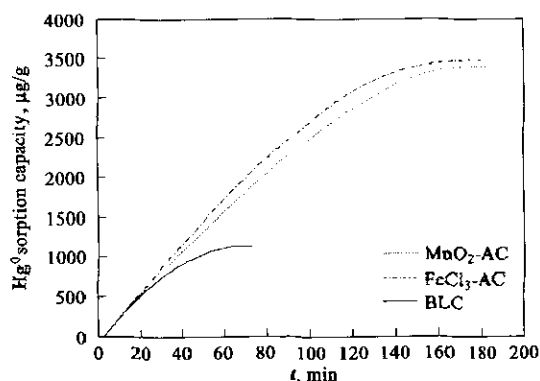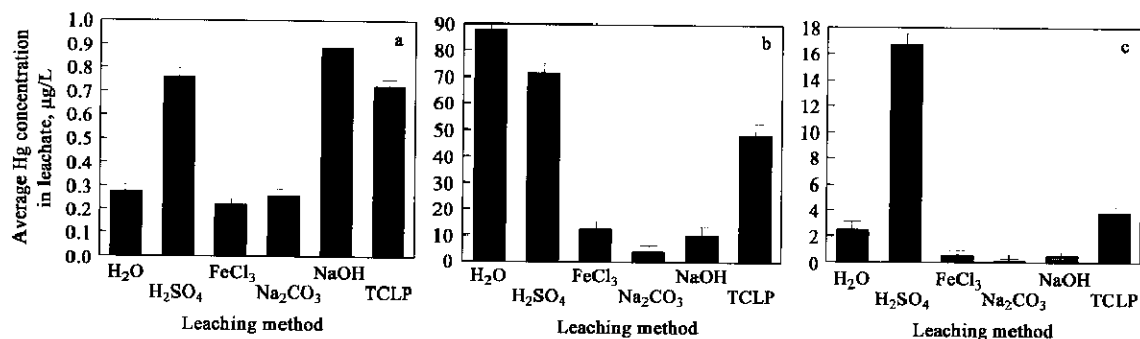
Fig.4   Hg leaching tests of BLC (a), MnO$_2$-AC (b) and FeCl$_3$-AC (c)

the surface of MnO$_2$-AC is not firmly held. As can be seen from Fig.4c, the average Hg concentration in each leachate for the FeCl$_3$-AC tests was low except for the 1% H$_2$SO$_4$ leachate. However, all the leaching results for the FeCl$_3$-AC tests are below the safe limit. Figs.4b and 4c show the Hg adsorbed by the treated ACs, especially by the MnO$_2$-AC, are not stable in the acid solution environment. The concentrations of Hg were relatively lower in the alkaline solution environment for both treated carbons. To clarify these facts, more researches should be done.

The conditions for Hg adsorption experiments were the same for the three ACs tested. However, it is clear that the amounts of Hg leaching out from the MnO$_2$-AC were much higher than those from the another two ACs as shown in Fig.5. There was very little Hg leaching out from the BLC relative to those from the MnO$_2$-AC and the FeCl$_3$-AC. It can be speculated that the oxidation treatment changed the physical and chemical characteristics of Hg on the surface of ACs. These changes induced the instability of Hg on AC in the solution environment.

Based on the discussions in Sections 2.1 and 2.2, there could be different forms of mercury such as Hg (NO$_3$)$_2$ and Hg$_2$(NO$_3$)$_2$ existing on the surface of MnO$_2$-AC due to the presence of MnO$_4^-$, Mn^{4+}, NO$_3^-$ on the

surface. Furthermore, both Hg (NO$_3$)$_2$ and Hg$_2$(NO$_3$)$_2$ are easily soluble in the water and acid solution (Lv et al., 1995). For FeCl$_3$-AC, the amount of Hg leaching out was between those from BLC and MnO$_2$-AC. It is speculated that Fe^{3+} may be the only possible oxidizing medium and Cl$^-$ was the primary anion on the surface of FeCl$_3$-AC. Therefore the oxidation potential of FeCl$_3$-AC may be weak compared to MnO$_2$-AC. Consequently, both HgCl$_2$ and Hg$_2$Cl$_2$ may be the most possible forms of Hg existed on the surface of FeCl$_3$-AC, and Hg$_2$Cl$_2$ could be the main Hg species. Although both MnO$_2$-AC and FeCl$_3$-AC have high Hg^0 sorption capacity as a result of oxidation treatment, however, Hg is much more stable on the FeCl$_3$-AC than the MnO$_2$-AC in solution environment because the Hg$_2$Cl$_2$ is slightly water soluble. Hence, it is very important to consider the stability of Hg on the surface of the sorbents after adsorption if using oxidation treatment to enhance the Hg sorption capacity of the AC sorbents.

2.4 Cost of treated AC used for Hg control

A good AC absorbent should have higher Hg adsorption capacity, better Hg adsorption stability, and lower cost. Based on the methods used for modifying AC in this research, the cost of oxidation treatment was 60%—70% of the production cost of BLC by an active MnO$_2$-solution, and was 40% —45% of the production cost of BLC by a FeCl$_3$-solution. For calculating relative cost of Hg control with oxidation treated ACs, the parameter C_R was used. Here, C_R is the Hg^0 sorption capacity per unit price of AC. It is assumed that the Hg^0 sorption capacity of BLC was 1 M$_{Hg}$/g. Then, the Hg^0 sorption capacity of MnO$_2$-AC, or FeCl$_3$-AC, is about 3 M$_{Hg}$/g. Similarly, it is assumed that the produced cost of BLC is 1 unit/g. Then, the production cost of MnO$_2$-AC is 1.60—1.70 unit/g, and that for FeCl$_3$-AC is 1.40—1.45 unit/g. The calculated C_R of BLC is 1 M$_{Hg}$/unit, C_R of MnO$_2$-AC is about 1.76—1.88 M$_{Hg}$/unit, and C_R of FeCl$_3$-AC was about 2.07—2.14 M$_{Hg}$/unit. The Hg^0 sorption capacities normalized to per unit cost of carbon production for the two treated ACs are higher that for the BLC. It is concluded that the cost of coal-fired flue gas Hg control would be decreased by using MnO$_2$-AC
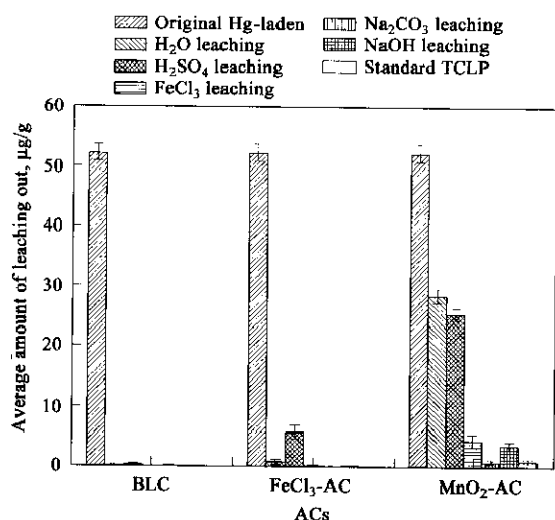

Fig.5 Results of Hg stability on the three ACs

instead of BLC. If FeCl₃-AC is used to replace BLC, the cost would even be lower.

3 Conclusions

The results of the surface analysis for BLC, MnO_2-AC, and $FeCl_3$-AC confirmed that the oxidation treatment by an active MnO_2-solution or a $FeCl_3$-solution on the surface of BLC not only changed the surface physical structure of BLC, but also modified the surface chemical composition of BLC with the addition of possible components such as MnO_4^-, Mn^{4+}, O, Fe^{3+}, NO_3^-, Cl^-. The results of the adsorption capacity experiments show Hg^0 sorption capacities of MnO_2-AC and $FeCl_3$-AC were about three times higher than that of BLC.

Based on the results of Hg adsorption stability on the surface of ACs experiments, it could be concluded that Hg was stable on the surface of the BLC in the solution environment. Although the average Hg concentration in the leachate of $FeCl_3$-AC was higher than that of BLC, all the leaching results of $FeCl_3$-AC were still less than the safe limit of 25 μg/L. However, the leaching results of MnO_2-AC show the average Hg concentrations in the leachates of H_2O and 1% H_2SO_4, and TCLP were over the safe limit. It is speculated that Hg existed most likely in the form of $Hg(NO_3)_2$, or $Hg_2(NO_3)_2$ on the surface of MnO_2-AC, and both of them were easily soluble in the water and the acid solution. For $FeCl_3$-AC, it is spectulated that Fe^{3+} may be the only possible oxidizing medium and Cl^- was the primary anion on the surface of $FeCl_3$-AC. Therefore $HgCl_2$ and Hg_2Cl_2 were the most possible Hg existing forms on the surface of $FeCl_3$-AC, and moreover, Hg_2Cl_2 should be the main species due to its low water solubility. It is very important to consider the stability of Hg on the surface of sorbents after adsorption if using oxidation treatment to enhance Hg sorption capacity of AC sorbents.

Based on AC absorbent's requirements of high Hg adsorption capacity, low cost, and high Hg adsorption stability, it can be concluded that the $FeCl_3$-AC is the best absorbent among the three ACs tested in this study.

References:

Brown T D, Smith D N, Hargis R A *et al.*, 1999. Mercury measurement and its control: what we know, have learned, and need to further investigated[J]. Journal of the Air Waste Management Association, Critical Review: 1— 97.

Kilgroe J D, Srivastava R K, 2000. Technical memorandum: control of mercury emissions from coal-fired electric utility boilers [M]. USEPA.

Karatza D, Lancia A, Musmarra D *et al.*, 2000. Study of mercury absorption and desorption on sulfur impregnated carbon [J]. Experimental Thermal and Fluid Science, 21: 150—155.

Luo Z Y, Hu C X, Zhou J S *et al.*, 2006. Stability of mercury on three activated carbon sorbents[J]. Fuel Processing Technology, 87(8): 679—685.

Lv Y Y, Wang W S, Liu S Y *et al.*, 1995. The sixth volume of inorganic chemistry series[M]. Bejing: Science Press.

Pavlish J H, Mann M D, 1998. An economic basis for developing mercury control strategies[R]. POWER-GEN International 1998, Orlando FL, USA.

Pavlish J H, Sondreal E A, Mann M D *et al.*, 2003, Status review of mercury control options for coal-fired power plants[J]. Fuel Processing Technology, 82: 89—165.Ren J L, 2003. Experimental and theoretical study on mercury transformation and sorbents adsorption in simulated combustion flue gases [D]. Ph. D Thesis. Zhejiang University, Hangzhou, China.

Shanghai activated carbon Co., Ltd., 2004. Technology data[EB]. http://www.shhxtc-carbon. com/info_03 _c. html (accessed August 18, 2004).

The Universal Treatment Standard [EB]. Accessed June 2004. http://www.epa.gov/ epaoswer/hazwaste/ldr/268_48.pdf.

USEPA, 2000. Performance and cost of mercury emission control technology applications on electric utility boilers [S]. EPA-600/R-00-083, National Risk Management Research Laboratory.

USEPA, 1997. U.S. EPA office of air quality planning and standards[S]. EPA-452/R-97-003. U.S. Government Printing Office: Washington, DC.

USEPA, 2002. Control of mercury emissions from coal-fired electric utility boilers[S]. EPA-600/R-01-109, National Risk Management Research Laboratory.

USEPA method 1311, 1992. Toxicity characteristic leaching procedure [EB]. http://www. epa.gov/epaoswer/hazwaste/ test/pdfs/1311. pdf (accessed June 2003).