

Comparison of Mercury Emissions in USA and China-The Way of Effective Control of Hg from the Power Plant

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Abstract

Coal-fired power plants are the largest sources of mercury in China as well as United States, accounting for nearly 50% of industrial mercury releases. USA consumed much more coal than China before 1987, while no Hg control measures had been taken during this period. There has been lots of background Hg emitted into air system, including those from mining, oil & gas extraction, coal combustion, volcanoes and geothermal et.al. However the coal combustion has been considered as the dominant sources. Since 2005, quite a few coal power plants started adopting mercury control measures based on the powder Active Carbon Injection (ACI) technology in USA, metallic Hg emission to air environment has been thus significantly reduced, but leading to Hg content in fly ash is up to 10 ppm, potentially a Hg pollution source if not disposed properly. Although short terms studies have showed little leachate of Hg occurred from the fly dust, there is a potential that the landfilled or dumped dust may be a possible source for the Hg leachate to contaminate underground water in long term, as Hg is mainly trapped as Hg²⁺ or other forms, which are either soluble or minor soluble in water and could go to water environment. Based on this analysis, a more safe and reliable Hg removal technology has been developed and tested.

Keywords: USA; China; Coal power station; Mercury removal

Introduction

Mercury is a heavy metal pollutant with high toxicity, volatility, persistence and bioaccumulation in the environment. Mercury compounds are emitted into the atmosphere from various anthropogenic and natural sources, and are later transported to the surface water and land. It has been shown that mercury content in coal varies between 0.01 and 1.5 g per ton of coal, with world coal consumption in 2011 estimated at 7695 million tons per year therefore coal consumption has been considered as one of the main resources of Hg emission [1].

According to UNEP, current anthropogenic sources are responsible for about 30% of annual emissions of mercury to air [2-6]. Since 2005, the increases in the application of air pollution controls in USA particularly the selective catalytic reduction (SCR), together with more stringent regulations in a number of countries have reduced mercury emissions from coal burning in power plants, and thus offset some part of the emissions arising from the increased coal consumption. In the United States, for example, emissions from coal burning at power plants have reportedly decreased from about 53 tons in 2005 to 27 tons in 2010. However, this is only based on the mercury vapor reduction rate from the coal power station, the reduced Hg was collected, which is in fact still mixed with the fly ash and exists in environment.

In the report of the Global Mercury Assessment 2013 (UNEP, 2013, Global Mercury Assessment 2013: Sources, Emissions, Releases and Environmental Transport. UNEP Chemicals Branch, Geneva, Switzerland), the total anthropogenic emissions of mercury to the atmosphere in 2010 were estimated at 1960 (in the range 1010-4070) tons which is about 30% of the total mercury that was emitted and re-emitted from anthropogenic and natural sources in that year. East and Southeast Asia are responsible for about 40% of global anthropogenic emissions. It is estimated that about 75% of the mercury from this region comes from China, which is about one-third of the global total.

Mercury is dangerous to aquatic and human life [1]. When mercury is deposited in lakes or streams, natural bacteria action converts it to

methyl mercury, which makes the mercury available to concentrate in the tissue of fish, wildlife and people who eat the fish. Humans, plants and animals are routinely exposed to mercury and accumulate in Hg containing environment, potentially resulting in a variety of ecological and human health impacts. Human exposure to mercury can result in long-lasting health effects, especially on fetal development during pregnancy. In addition, mercury poisoning has been linked to nervous system disorders, kidney and liver damage, and impaired childhood development.

In a recent bilateral environmental meeting between China and USA in 2006, the director general of US EPA, Dr. Steven Johnson from EPA pointed out that the US Hg contamination may come from China. In Dec 2013, the US EPA director Gina McCarthy said that the Environment in USA West Coastal was being affected by the air pollutant migrated from China. It is assumed that Hg emitted from China migrated in air environment and deposited to the agriculture and the rivers in USA.

This saying has been there for some time, and so far, there have been no direct data on the US and China Hg emission and also no deep understanding on the control way of Hg to effect on agriculture and rivers. In this work, we review the Hg emission from coal power station, and the effectiveness of the Hg control ways, so as to give a more reliable insight into the Hg emission in the two countries.

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Received January 29, 2014; Accepted March 24, 2014; Published April 01, 2014

Citation: Chen H, Wang X, Zhong L, Xu S, Xiao T (2014) Comparison of Mercury Emissions in USA and China-The Way of Effective Control of Hg from the Power Plant. Int J Waste Resources 4: 138. doi: [10.4172/2252-5211.1000138](https://doi.org/10.4172/2252-5211.1000138)

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Method and Experimental Setup

In this work, the coal consumption and the emission status in the past years have been reviewed with the focus is on the US and China, including coal consumption, the control measures and the final treatment of the Hg containing materials, based on the open available literatures.

We also report a novel Hg sorbent, which has been developed and tested using a fixed bed micro-reactor test system, the mercury is brought into the sorbent-loaded tubular reactor, the metallic Hg content inlet and outlet were analyzed using an Hg analyzer. The unabsorbed Hg in the vent is absorbed with HNO_3 - H_2O_2 and KMnO_4 solution. The schematic setup of the Hg test is shown in Figure 1.

In this test setup, a cold vapor atomic fluorescence spectrometry mercury vapourmeter MODELIII made by Brooks Rand was used to detect the concentration of gaseous state elemental mercury. It can measure metallic mercury concentration in the gas stream down to ppt level. In order to ensure the measured value and set value perfectly, mercury concentration fluctuation less than $\pm 2\%$ hourly was required. Gas supply part used the mass flowmeter to control all kinds of gas flow in mixing for simulating flue gas composition. Water vapor and simulated flue gas flows through the quartz tube from inner pipe and outer tube respectively, and was mixed in the fixed bed upper part, then was absorbed by fixed-bed sorbent reactor. After the reaction, gas passed through the mercury measurement instrument which measured the metallic mercury in flue gas. The whole pipe and pipe connection direct connect to mercury using PTEF to protect the surface inside from Hg0 adsorbed in our experimental system. The Hg sorption test was carried out at 140°C , with linear velocity in 0.5-5 m/s.

XRD measurement of the spent sorbent was carried out using X-ray diffraction(XRD) with a Rigaku D/max 2550 diffractometer using CuK_α radiation ($\lambda=0.1541\text{ nm}$) over a 2 theta range from 10° to 80° at a 0.02 step, so as to identify the Hg phase in the sorbent. The Hg pick up capacity is determined by the weight change of the sorbent as well as the penetration curves.

Results and Discussion

Literature results of the re-emitted Hg in the environment

According to the UNEP investigations, current anthropogenic sources are responsible for about 30% of annual emissions of mercury to air [2-6]. Another 10% comes from natural geological sources, and the rest (60%) is from 're-emissions' of previously released mercury that has built up over decades and centuries in surface soils and oceans, although the original source of this reemitted mercury cannot be detected.

However, it has shown that coal power plant accounts for nearly 50% of anthropogenic Hg emission. Taking the coal consumption changes with the time, as shown in Figures 2 and 3, the coal consumption graph from 1965 to 2012 showed that before 1987, US used much more coal than China, and there had no Hg removal measures or action taken at that period, so it is inferred that a fairly amount of the re-emitted mercury in the early stages was from USA, rather than from China.

The coal consumption in China started to exceed USA from 1987, and the exceeded amount is about 100Mt of coal from 1995 to 2004. Thereafter, China was expected to emit more mercury than USA in this period, but the net surplus Hg emission from China is less than the Hg amount emitted from USA before 1987 when considering the coal consumption amount and assumed not Hg removal measures in place.

The coal consumption in China increased sharply from 2004 onwards, and peaked at 1.9 billion tons in 2012 [1,7-9]. The Hg emission from China in this period is the largest in the world. As shown by Jiming Hao et al. [1,10], the Hg emission in 2008 in China may range from 57 tons to 183 tons depending on the confidence interval. However, the migration of metallic Hg is slow and may be most bonded to the local dust and soil, which is easier to contaminate the local environment. Therefore, even the transport distance of the metallic Hg is short, China should pay more attention to the power station locations, as the Hg may mostly deposit in nearby. To prevent the soil from pollution, Hg purification technologies and facilities are urgently needed to install in China, as the coal consumption is soaring since 2005, even the Hg will

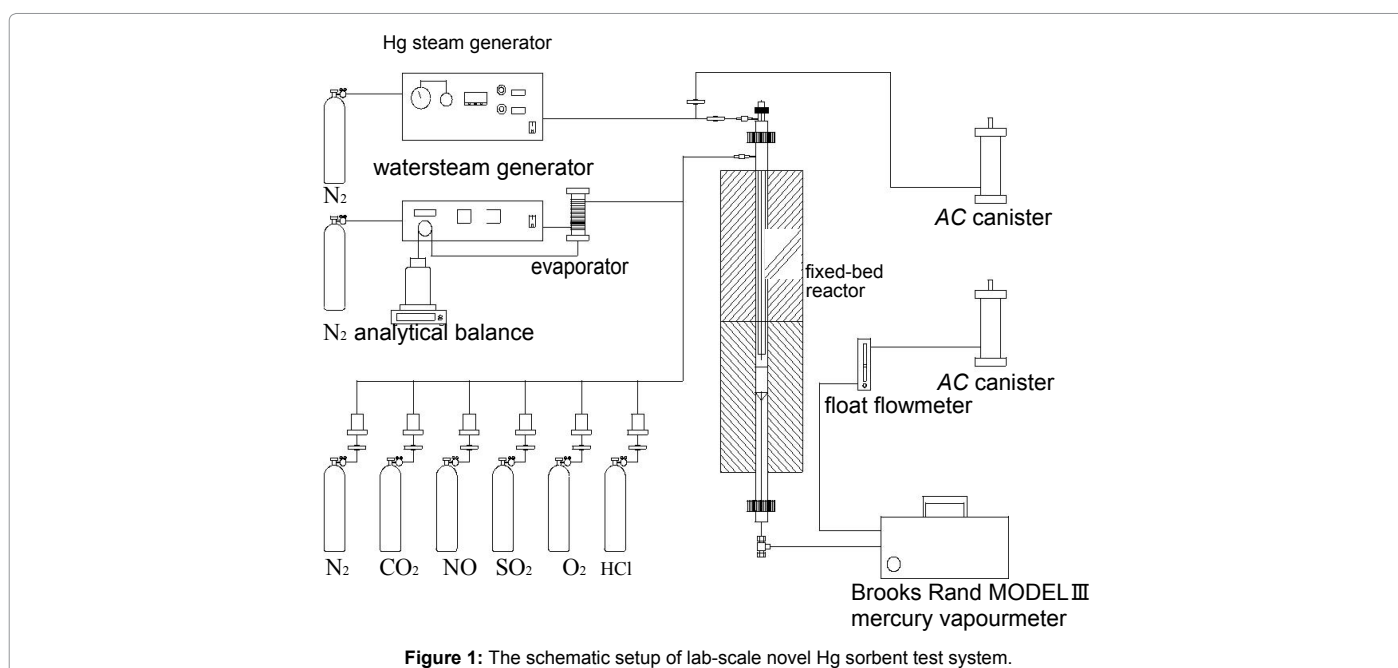
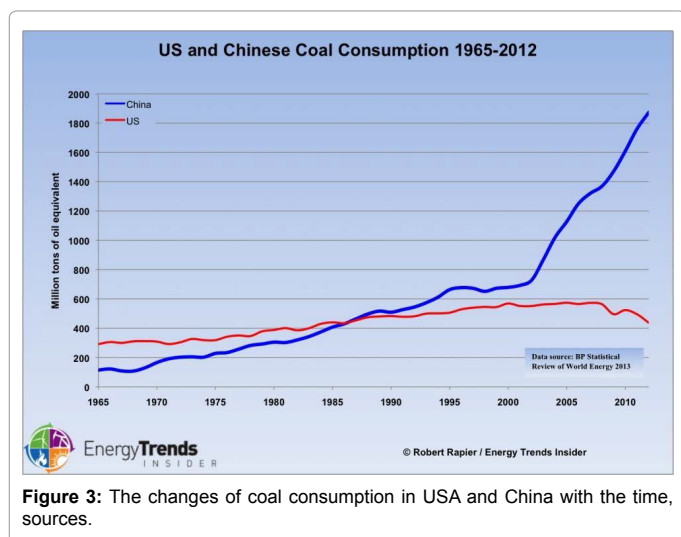
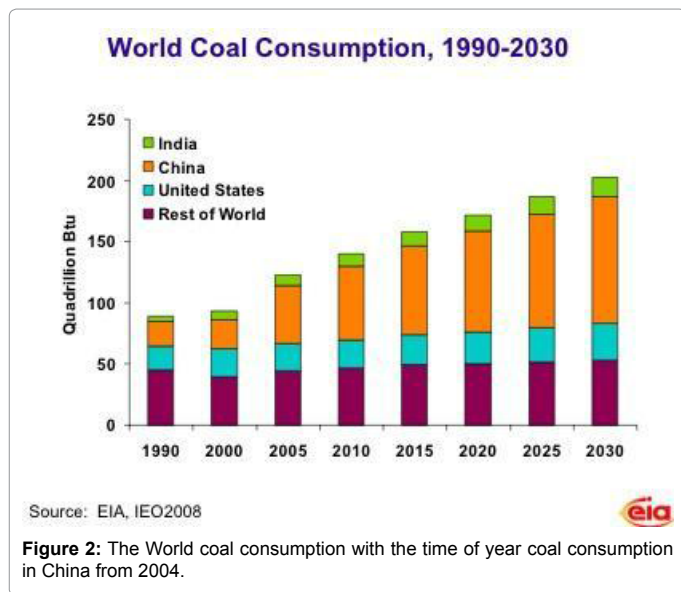


Figure 1: The schematic setup of lab-scale novel Hg sorbent test system.



not migrate to USA, the deposited Hg may contaminate the soil and water in China, which goes into the food chain, and in the end affect the whole world [1,11-15].

Mercury control technology and the remaining issues

The pollution of mercury emission has caused worldwide attention, and many country governments have set out regulations to control Hg emission [16-20]. The US EPA firstly regulated newly built or reconstructed cement plants Hg emission in 2002, and required them to install Hg removal after December 2, 2005. In 2013 U.S. EPA set new limits on mercury emissions for the coal power plants, the Hg emission should be less than 1.36×10^{-3} kg/GWh for all the power stations in 2015. This emission limits correspond to 10 and 4 $\mu\text{g}/\text{Nm}^3$, which are the strictest mercury emission limits. Before that, the mercury removal was mostly removed using the activated carbon injection system, which is shown in Figure 4.

The US is making significant effort in decreasing harmful Hg emissions into the environment through regulations such as the EPA's Clean Air Interstate Rule (CAIR) and the Clean Air Mercury

Rule (CAMR), which placed caps on NO_x, SO_x and Hg emissions. In March 2011 the EPA stated that all hazardous air pollutants must have emission standards and proposed that for existing sources in the category, that the standards are at least as stringent as the emission reductions achieved by the average of the top 12% best controlled sources for source categories with 30 or more sources. With this new rule, a reduction of mercury from coal emissions of approximately 90% is anticipated continuously to improve.

Typically, activated carbon injection is used to capture oxidized mercury [21-32]. A major limitation with using activated carbon is that in flue gases with low halogen concentrations a large amount of activated carbon needs to be added to the system to effectively control Hg⁰. Over the last 10 years, most research and development work has focused on the full-scale and slip-stream field testing of activated carbon injection (ACI) and flue gas desulfurization enhancements at nearly 50 U.S. coal-fired power plants. The goal was to demonstrate high levels (50% to 90%) of mercury capture over an extended period of operation, while also reducing the cost of mercury removal. Until 2008, nearly 90 full-scale ACI systems have been ordered by U.S. coal-fired power generators, accounting for over 44 GW of coal-fired elec. generating capacity, and now more coal fired power plants are installing or considering installing the Hg removal facilities using activated carbon injection technology [33-35].

It should be noted that the injected carbon would absorb some of the metallic mercury and may convert it into Hg²⁺ when there exist halogen promoters, such as Br or Cl [36-40]. This is the reason lots of chlorinated or bromide activated carbon has been in the industrial application [41-44]. As shown in Figure 4, the Hg deposited carbon would be collected together with the fly ash. These Hg enriched ashes would be used for cement or land filled. Although there have been some studies on the potential effect of the Hg containing fly ash leachate. However, the leachate experiments were studied for a relatively short period, and so far, there is no quantitative estimation of the Hg in the coal. Here we make a rough estimation in the following.

Generally Hg content is 0.01 and 1.5 g per ton of coal, averaged at 0.17 g per ton [15,16,45,46]. The coal normally contains 5 to 24% of ash, in which average ash content is supposed to be 10%, and 80% of the ash is fly ash. Assume 80% of the mercury is captured by the active carbon, whose content in the collected ash may be neglected, the total mercury from each ton coal combustion in the collected fly ash can be calculated as follows:

$$\text{The total Hg collected } \text{MHg} = 0.8 \times 0.17 = 0.136 \text{ g}$$

The total ash collected from each ton of coal = $10\% \times 80\% \times 1000 = 80$ Kg of ash
The Hg content in the ash $0.136/80000 = 1.7$ ppm.

It should be noted that the Hg content in coal ranges from 0.01 g to 5.0 g in each ton of coal, but in the above calculation, Hg content in coal was set to be 0.17 g, and the Hg content in the resultant collected ash is 1.7 ppm. If Hg content is 1.7 g in coal, the resultant ash may contain up to 17 ppm. Therefore the Hg in the collected ash is in a wide range.

It is clear that the Hg in the collected ash is about 1.7 ppm even with 0.17 g Hg/ton of coal as the fuel, which is significant, thus the collected ash should be carefully disposed. In fact, quite a lot of this kind ash has been landfilled or stored in ash pounds, a short term leachate study may not be able to show the true changes the trace elements changes. There have been few reports on long period leachate study. As shown

How Power Plants Are Reducing Air Emissions

This simplified diagram is illustrative of the operations at a large coal-based electric power plant. It explains the various control technologies in place at many U.S. power plants to reduce emissions to air, land, and water. These technologies are designed to control emissions of nitrogen oxide (NO_x), sulfur dioxide (SO₂), and particulate matter (PM). In addition, these control technologies capture significant amounts of other air emissions, including mercury. The diagram also illustrates the ways that byproducts of coal combustion are recycled into useful products.

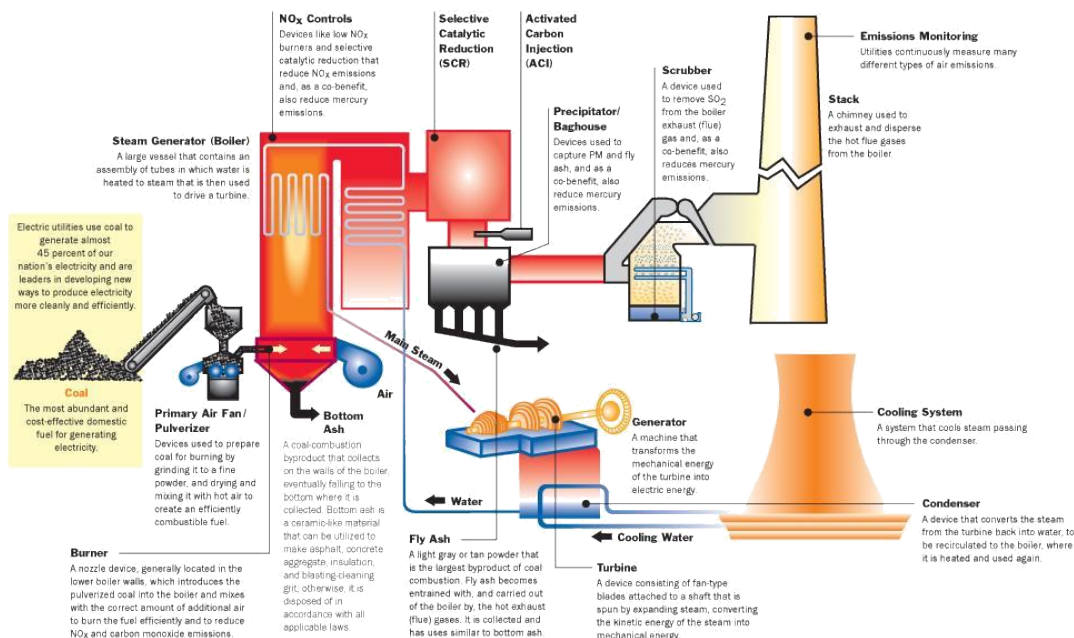


Figure 4: The typical flue gas purification schematic in US coal power plant.

	Name	Molar Weight (g/mol)	Melting point (°C)	Boiling point (°C)	Decomposition /sublimate temperature (°C)	Density (g/cm ³)	Aqueous solubility (g/l at 25°C)
Hg(0)	Elemental mercury	200.59	-38.8	356.7	n.a.	13.534	5.6 × 10 ⁻⁷
Hg ₂ Cl ₂	Mercurous chloride	472.09	525	n.a.	383	7.15	0.002
HgCl ₂	Mercuric chloride	271.5	277	302	n.a.	5.43	28.6
Hg ₂ SO ₄	Mercurous sulphate	497.24	n.a.	n.a.	n.a.	7.56	0.51
HgSO ₄	Mercuric sulphate	296.66	n.a.	n.a.	450	6.47	Decomposes
HgS	Mercury sulfide	232.66	n.a.	446-583	580	8.1	Insoluble
HgO	Mercuric oxide	216.59	n.a.	356	500	11.14	Insoluble
Hg ₂ Br ₂	Mercurous bromide	560.99	405	n.a.	340-350	7.307	3.9 × 10 ⁻⁴
HgBr ₂	Mercuric bromide	360.44	237	322	n.a.	6.03	Slightly soluble
Hg ₂ I ₂	Mercurous iodide	654.98	n.a.	n.a.	140	7.7	Slightly soluble
HgI ₂	Mercuric iodide	454.4	259	350	n.a.	6.36	0.06
Hg ₂ F ₂	Mercurous fluoride	439.18	n.a.	n.a.	570	8.73	Decomposes
HgF ₂	Mercuric fluoride	238.59	645	650	645	8.95	Soluble, reacts
Hg ₂ (NO ₃) ₂	Mercurous nitrate	525.19	n.a.	n.a.	70 (dihydrate)	4.8 (dihydrat)	Slightly soluble, Reacts
Hg(NO ₃) ₂	Mercuric nitrate	324.7	79	n.a.	n.a.	4.3	Soluble
Hg(CN) ₂	Mercuric cyanide	252.63	320	n.a.	n.a.	3.996	Soluble

Table 1: Properties of selected mercury compounds [20,47-49]. n.a.: not available.

in Table 1 of the various Hg compounds, HgCl₂ or HgBr₂ are either soluble or minor soluble in water, hence it may gradually leach out and pollute the environment if they are present in the collected ash.

Based on the above analysis, it is very difficult to conclude that the mercury collected in the injected activated carbon in US power plants has been securely stored and no secondary pollution of the collected active carbons occurs. Because if mercury is emitted from the flue gas in metallic form, it may transport into air environment. However,

when it is enriched as Hg²⁺ in the fly ash, it may increase the risk of the Hg leachate under some special conditions; more attention needs to be paid to the nearby underground water in the ACI Hg collection power plant.

Identification of the Hg species from the spent sorbent

So far in the fly ash collected from the active carbon injection technology, the Hg species in the sorbent is very difficult to be identified,

as the amount of Hg is too little to be detected. Here we have developed series Hg sorbent and tested them over a fixed bed test system.

The Hg break curves are shown in Figure 5, the Hg removal ratio is about 99.9999% in the first 17 days. The inlet Hg content is about 16806 ppb, with the Hg conversion about 99.9999%; the outlet Hg content is reduced down to lower than 0.1 ppb in a single pass. The Hg pickup capacity is up to 10 wt% according to the mass change of the sorbent. The spent sorbent was characterized using XRD and the results are shown in Figure 6. It is shown that the Hg is mainly present as HgCl₂, HgCl when CuCl₂ is present on the sorbent. For the bromided sorbent, Hg exists as HgBr₂. The SO₂ sulfur seems to have little influence on the formation of HgBr₂.

Because the Hg capacity is up to more 10 wt%, and given the Hg amount in a power plant is limited, it is proposed to install fixed bed Hg removal system in coal power station, e.g., installing a mercury removal sorbent bed in the chimney and let the flue gas flow through the sorbent bed after the dust is removed. The spent sorbent could thus be collected and properly landfilled, so as to avoid the secondary pollution by the leachate, which may be more difficult to remedy in the future.

Conclusion

Mercury emission from anthropogenic sources has become increasing serious issue and caused environmental pollution. More

than 60% of current Hg pollution might be from the re-emission of the Hg vapor discharged to the environment before 1987.

Coal fired power stations contribute to up to 40% of Hg emission worldwide. Before 1987, US consumed much more coal than in China, which may be the major source for Hg emission in the air, as no control measures were taken at that period.

In recent years, many countries started to regulate the Hg emission from coal fired power station and the measure is mainly based on activated carbon or halogen modified activated carbon injection. This can remove up to 90% of Hg from the flue gas, and converted it into Hg²⁺ and deposits over the carbon. However the Hg containing activated carbon is present in the fly ash in which Hg content can be up to 17 ppm, which may be a static source for Hg leachate if the fly ash is not disposed properly.

Novel Hg sorbent has been developed and tested in a fixed bed Hg removal micro-reactor whose Hg pick capacity is up to 10wt%, which can reduce the Hg in the flue gas to 0.1 ppb. The concentrated Hg sorbent has small volume and can be safely disposed easily.

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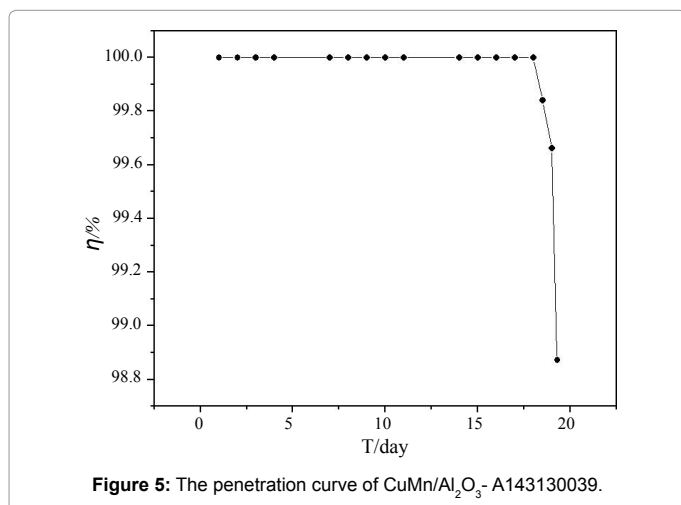


Figure 5: The penetration curve of CuMn/Al₂O₃ - A143130039.

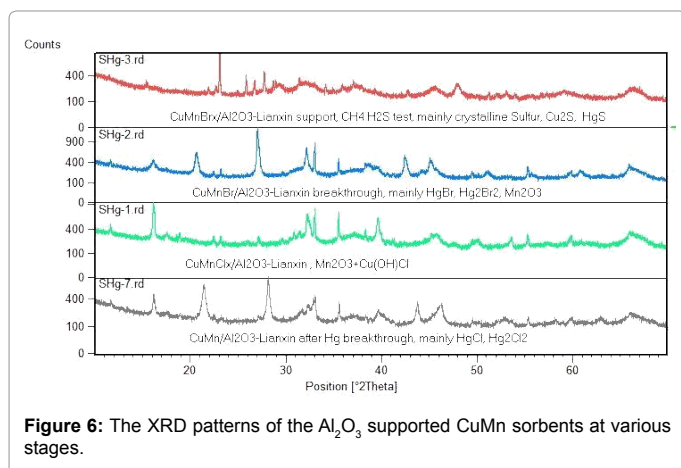


Figure 6: The XRD patterns of the Al₂O₃ supported CuMn sorbents at various stages.

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