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FUEL PROCESSING TECHNOLOGY

Activated carbon injection in spray dryer/ESP/FF for mercury and toxics control

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Abstract

In the last decade, dry scrubbing by spray dryer absorption (SDA) has been applied to more than 17 000 MW of coal-fired boilers, and several hundred municipal solid waste (MSW) and hazardous waste plants. In the MSW application, process development for air toxic control has advanced to a point where a spray dryer absorber, followed by a fabric filter, has been determined to be the best available control technology (BACT) in several countries throughout the world. Extremely high removal of dioxins and furans, as well as mercury and other trace toxics, is now state of the art in this application. The paper describes the recent development in the air toxics removal capability of dry scrubbing for coal-fired utility plants. Results of a study characterizing the inherent mercury removal efficiency of full-scale dry scrubbers in Europe and the United States are presented, as well as results of activated carbon injection tests performed at one full-scale installation. The paper further reports results of two pilot plant research programs. One pilot plant was equipped with an electrostatic precipitator, the other pilot plant with a baghouse. The paper reports results of activated carbon injection and identifies the parameters and conditions important for achieving high mercury removal in a dry FGD system. Options for control of mercury on eastern and western US coals are suggested. Results of mercury speciation are also given. Removal results of other vapor phase toxic trace metals, such as boron and selenium, are presented. Finally, the paper makes reference to the results of two mercury revolatilization studies.

1. Introduction

The SDA process combines spray dryer technology with efficient particulate collection in either an electrostatic precipitator or a fabric filter. The system is designed to

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remove acid gases and particulates from flue gases without saturating the flue gas, thereby producing a dry product for disposal. Fig. 1 shows a simplified diagram of the SDA process [1].

For additional control of vapor phase emissions such as mercury and dioxins, the SDA system can be augmented with an activated carbon injection system [2]. This consists of a system for injection of additives, preferably upstream of the spray dryer absorber.

Joy/Niro has been the leader in the development of spray dryer absorption technology for municipal solid wastes (MSW) and hazardous waste applications, and has spearheaded the development of process modifications that enable high removal of mercury and dioxins [3–7]. Significant experience has been gained in the removal mechanisms for trace toxic pollutants including heavy metals, dioxins, and organics [8–13].

The experience gained with mercury removal in MSW applications is summarized in Fig. 2. At the typical spray dryer outlet temperature in MSW applications of 260–280°F, the inherent mercury removal in the spray dryer absorber system is only 30–50%. By active carbon addition to the flue gas the removal of mercury can be increased to more than 90%. It is interesting to note that no significant difference in the mercury removal between an electrostatic precipitator and baghouse has been found.

2. Control of mercury from coal-fired combustors

2.1. Mercury removal in US/European dry FGD systems

Recently, Joy/Niro has measured the inherent mercury removal efficiency achieved in a number of operating SDA systems on coal-fired power stations, both in the US and in Europe [15]. Results of the characterization of mercury removal efficiency at eight dry scrubbing installations are shown in Table 1. The mercury concentrations

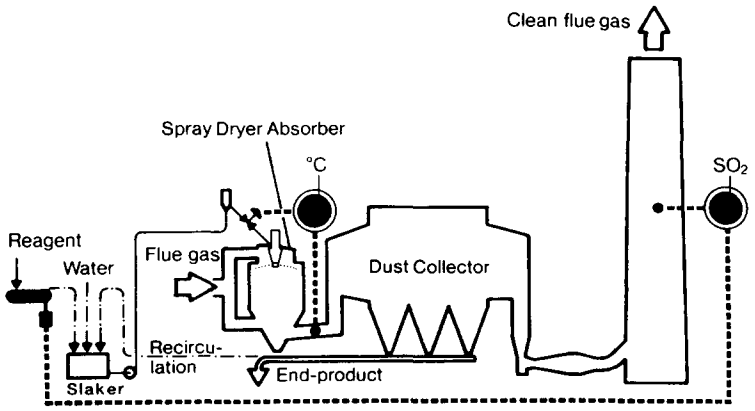

Fig. 1. Spray dryer absorber process.

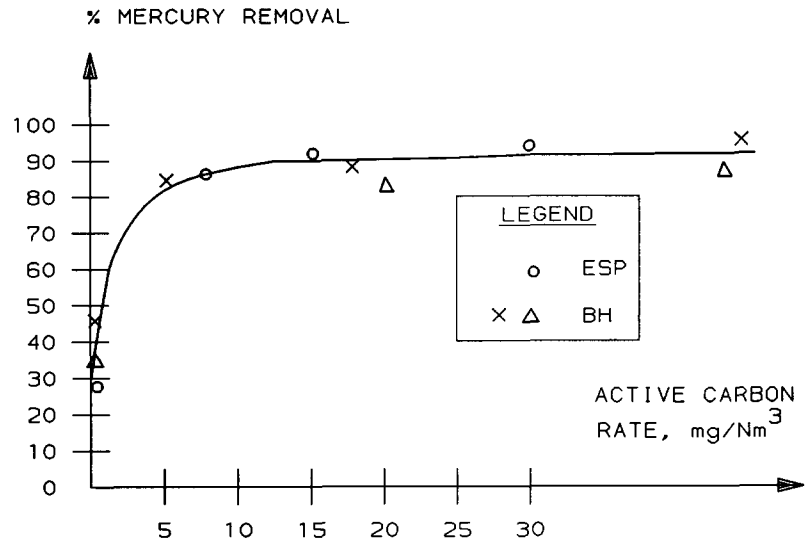

Fig. 2. Effect of active carbon addition on mercury removal for MSW ($T = 280°F$).

Table 1
Mercury removal in full scale dry FGD systems

Plant	Coal type	Particulate collector	Active carbon injection	Mercury concentration (μg/N m³)		% Mercury removed
				SDA inlet	Stack	
A	Western US	BH	—	10.5	9.1	14
B	Western US	ESP	—	4.3	3.3	23
C	Western US	BH	—	8.4	7.9	6
D	Eastern US	BH	—	5.1	0.18	96.5
D	Eastern US	BH	+	3.9	<0.03	>99
E	Columbian	BH	—	2.9	1.3	55
F	Polish	ESP	—	2.7	0.31	89
G	Western US	BH	—	6.8	4.2	16
H	Spot Market	BH	—	6.3	3.5	44

indicated are the vapor phase mercury only. Previous investigations [16] have shown that more than 99% of the mercury emitted from a coal-fired power station is present as volatile vapor phase mercury. Depending on the coal type, the uncontrolled mercury concentration in the flue gas was found to be in the range 35–11 μg/N m³. The indicated mercury concentrations have been corrected to 5% oxygen. As can be seen from the table, the inherent mercury removal efficiency ranges from a low of 6% to a high of 96%. In the eastern US installation (Plant D), the mercury removal efficiency was further enhanced by active carbon injection upstream of the spray

Table 2
Mercury removal parameters

Plant	Fly ash loading to SDA	Coal chlorine content	% Mercury removed
A			14
B			23
C	High	Low	6
G		(<0.01%)	16
E	Low		55
H			44
F	Medium	High	89
D	High	(0.1–0.3%)	96

dryer absorber. This reduced the outlet emission an order of magnitude and increased the removal efficiency to more than 99%.

As mentioned above, the previous experience with SDA systems installed on MSW plants has shown that active carbon injection into the flue gas upstream of the spray dryer absorber has a significant influence on the removal efficiency of mercury. It would therefore be reasonable to expect that active carbon injection also could be used to enhance the removal efficiency on coal-fired applications. If active carbon is not injected, one would expect that the fly ash loading and the LOI would be of significance for the mercury removal. In Table 2, the data in Table 1 have been rearranged to show the correlation between mercury removal and fly ash grain loading as well as coal chlorine content. From the numbers in Table 2 it seems certain that the mercury removal efficiency can be correlated with the fly ash grain loading. The table further indicates that a strong influence of the coal chlorine content exists. From Tables 1 and 2, it further appears that the mercury removal efficiency in a spray dryer system is independent of whether an electrostatic precipitator or a baghouse is used as dust collector.

2.2. *Results of SH pilot plant research program*

The mercury removal results achieved in the full-scale dry scrubbing systems described above clearly indicate that the dry scrubbing system has the capability of achieving high mercury removal efficiency. To further investigate the air toxic removal capability of the dry scrubbing process, a pilot plant research program was funded by Niro in cooperation with the Sonderjyllands Hojspaendingsvaerk (SH).

A pilot plant was installed on a slip stream of a 600 MWe boiler at the SH Power Station in the southern part of Denmark. A representative sample of flue gas was passed through a pilot spray dryer absorber and an electrostatic precipitator. The size of the pilot plant was 2 MWe corresponding to 7000 $N m^3/h$. The pilot plant was equipped with an activated carbon injection system upstream of the spray dryer absorber. The plant was operated in the normal recycle mode of operation, which is

Table 3
Coal analysis – SH pilot plant

	Coal A		Coal B
Type	COCERR, Columbia		US Mix 35% US West 65%
Heating, value, Btu/lb	6450		6690
Analysis, wt%	C	67.1	68.4
As received	H	4.4	4.3
	N	1.4	1.4
	S	0.86	1.05
	Cl	0.019	0.094
	Ash	8.4	9.6
	H_2O	12.4	10.5
Mercury, ppm	0.073		0.088

typical for spray dryer absorber systems. The operation was steady during the tests and an average of 90% SO_2 removal was achieved. The plant operated at 20°C approach to saturation temperature.

During the test program two types of coal were burned. Table 3 shows the characteristics of the two coals. It is interesting to note that the chlorine content in coal B is 5 times higher than in coal A. The mercury content of the coal resulted in flue gas concentrations entering the spray dryer absorber of 5–8 $\mu g/N\,m^3$.

The pilot plant was at first operated at baseline conditions with no active carbon injection. Fig. 3 shows that at baseline conditions, 57% removal was achieved with coal A and 78% removal was achieved with coal B. When active carbon was injected into the flue gas, removal efficiency of mercury could be increased for both coal A and coal B to more than 90%. It is, however, interesting to note that 90% removal of mercury with coal A requires 5 times as much active carbon than with coal B. As documented below, this is due to the difference in mercury speciation as a result of the different coal chlorine content.

During the whole test program, speciation of mercury inlet to the spray dryer absorber and outlet of the electrostatic precipitator was performed. Table 4 shows mercury speciation data. Each test is an average of three samplings. Sampling inlet to the spray dryer absorber and outlet of the precipitator was done simultaneously. As can be seen from the speciation data, the major part of the mercury inlet to the spray dryer absorber is present as oxidized mercury, most likely in the form of mercury chloride. Fig. 3 shows a plot of the data from Table 4. The figure is clearly showing the effect of carbon injection on elemental and oxidized mercury removal, respectively. With no carbon injection, the inherent removal of elemental mercury in the SDA system is virtually zero. In contrast, the inherent removal of oxidized mercury is 95%. With active carbon injection, removal of elemental mercury can be increased from 0 to approximately 50–60%. Hence, the overall removal of mercury will depend on the speciation of mercury inlet to the spray dryer and the quantity of active carbon added to the flue gas.

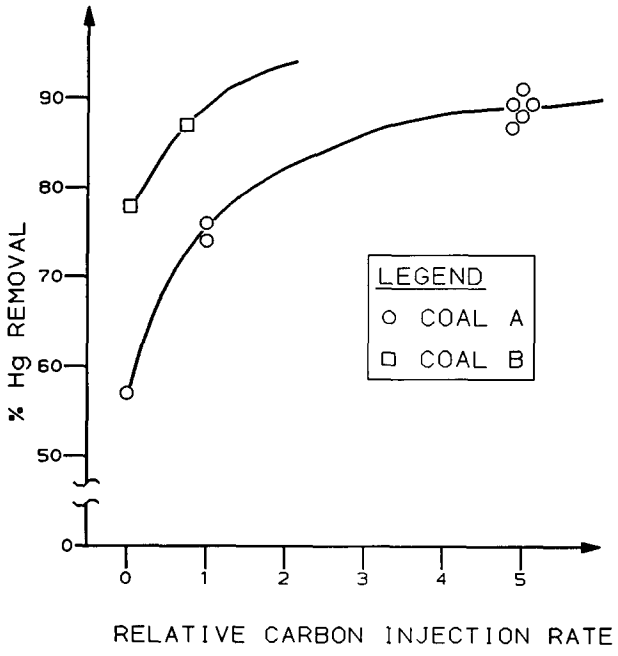

Fig. 3. Mercury removal results — SH pilot plant.

Table 4
Mercury speciation data (μg/N m^3) – NSP pilot plant

Coal	Test no.	Carbon rate	SDA inlet		FF outlet	
			Hg^{2+}	Hg^0	Hg^{2+}	Hg^0
C	13/1		0.70	2.36	0.03	2.63
C	13/2	0	0.96	3.07	0.06	2.82
C	14/2		1.05	2.60	0.14	3.12
W	04/1	1	1.15	4.82	0.58	3.53
W	06/1	5	2.01	6.52	0.32	3.68
W	06/2		1.94	6.64	0.39	3.99

2.3. Results of NSP, Sherco pilot plant research program

Further research efforts to study mercury emissions from coal-fired power plants were carried out at Northern States Power, Sherco 3 Station. Cypress Energy Partners (CEP), Northern States Power Company (NSP), Joy Environmental Technologies, Inc. and Niro A/S have formalized a testing agreement under which the feasibility of removing mercury from coal-fired power plant flue gases was investigated.

The test program was conducted at Northern States Power, Sherburne County Generating Plant, located in Becker, Minnesota. The flue gas from Sherco Unit 3, an

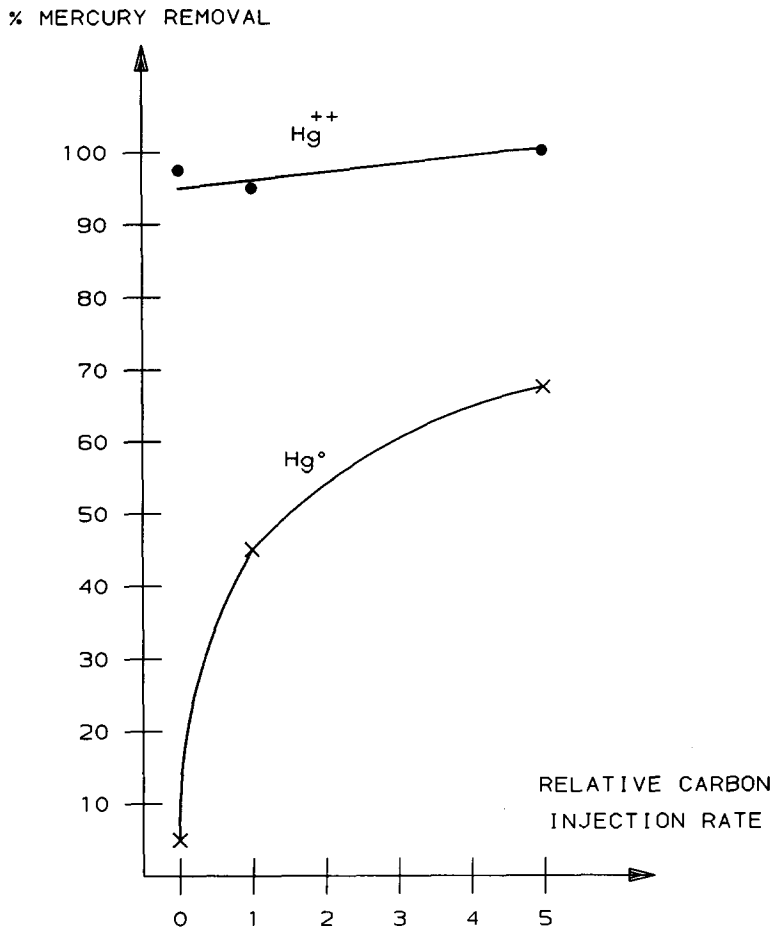

Fig. 4. Effect of carbon injection on Hg^0 and Hg^{2+} removal ($T = 160°F$).

800 MWe coal-fired steam electric generating facility equipped with a spray dryer absorber and baghouse, was directed to a pilot plant via a slipstream elbow positioned in the ductwork upstream of one SDA module. The pilot plant consisted of an SDA and baghouse capable of handling 300 N m^3/h of flue gas. The pilot plant was fully equipped with feed preparation and recycle system. During the mercury testing, the pilot plant was run at exactly the same conditions as the full-scale dry scrubbing installation.

To validate the pilot plant results, a base test was run, where inlet and outlet mercury concentrations of both the full-scale dry scrubbing system and the pilot plant were simultaneously measured. Mercury removal in the pilot plant and the full-scale SDA system agreed well, and for the base condition in the order of 10–20%. After the baseline test was performed, a number of tests with injection of normal activated carbon upstream of the spray dryer absorber were carried out. Fig. 4 represents the

Table 5
Mercury speciation data (μg/N m^3) – SH pilot plant

Test no.	Carbon rate	Inlet SDA		Outlet ESP	
		Hg^{2+}	Hg^0	Hg^{2+}	Hg^0
0		6.03	1.94	0.21	1.60
1	0	6.46	1.29	0.22	1.10
5		3.03	1.52	0.08	1.86
2		5.36	1.54	0.35	1.01
3		5.98	2.54	0.27	0.94
12	1	3.50	1.67	0.05	1.20
13		3.10	1.72	0.16	1.07
8		2.99	1.51	0.03	0.38
9		2.83	0.88	0.01	0.38
10	5	3.33	1.26	0.01	0.48
11		3.83	1.52	0.03	0.68
14		2.20	1.57	0.07	0.46

mercury removal efficiency achieved as a function of active carbon injection rate. By active carbon injection, the mercury removal efficiency could be increased to the 50–60% level. This level was far from the targeted removal efficiency of 90%. Swedish investigations [17,18] have shown that mercury reacts with HCl or chlorine at moderate temperatures in the presence of activated carbon. Therefore, a number of tests using either HCl or chlorine injection into the flue gas immediately upstream of the spray dryer absorber were performed. As can be seen in Fig. 4, neither HCl nor chlorine injection improved the removal of mercury significantly. It is speculated that the added HCl and chlorine reacts with the alkaline fly ash, thereby being inactivated and not available for reaction with the mercury.

The reason for the relatively low mercury removal achieved by active carbon injection can be found by analyzing the speciation data in the inlet to the spray dryer absorber and the outlet of the fabric filter. These data are presented in Table 5. The major part of the mercury present in the flue gas was elemental mercury. Only 10–15% was present in the oxidized form, and is most likely present as mercury oxide. By the active carbon injection, up to 80% of the oxidized mercury could be removed, whereas the elemental mercury removal was 40–50%.

Chemical reaction equilibrium would indicate that iodine and sulfur react with elemental mercury. Therefore, tests using iodine-impregnated and sulfur-impregnated activated carbon were performed. In Fig. 5, the significant improvement achieved by using iodine- and sulfur-impregnated carbon can be seen. With the iodine-impregnated carbon, virtually 100% removal of mercury could be achieved. With sulfur-impregnated carbon, more than 80% removal efficiency was reached.

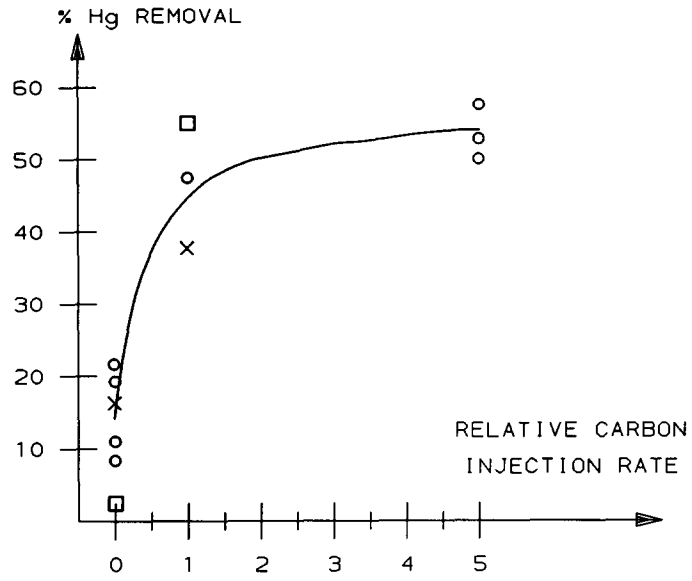

Fig. 5. Mercury removal by active carbon injection NSP pilot plant. (○) Normal activated carbon; (×) Cl_2 injection; (□) HCl injection.

2.4. Discussion of results

The pilot plant research work reported above, as well as the full-scale measurements, leads to the conclusion that a dry scrubber operating at close approach to adiabatic saturation has the capability of very high removal of oxidized mercury (95%) without using activated carbon injection. In contrast, elemental mercury is passing through the system untouched. However, by using activated carbon injection, elemental mercury can also be captured. It is interesting to compare the removal efficiency of oxidized and elemental mercury in a dry scrubber with similar results for a wet scrubber, recently published by German investigators [19]. Fig. 6 shows the result of their measurements on full-scale wet scrubbers. It also shows the distribution of elemental and oxidized mercury based on inlet conditions to the FGD system. From the figure, it can be seen that more elemental mercury is leaving the scrubber than entering it. This is quite remarkable and is believed to be due to a conversion of some of the captured oxidized mercury back to elemental mercury in the wet scrubber. The converted elemental mercury is revolatilized into the flue gas. This, of course, decreases the overall mercury removal efficiency in the wet scrubbing system.

The research results seem to indicate that the chlorine content of the coal has a major impact on mercury removal. With chlorine present in the coal, part of the mercury will exit the boiler as mercury chloride. Based on the performed full-scale and pilot-scale measurement, an attempt to quantify the correlation between coal chlorine

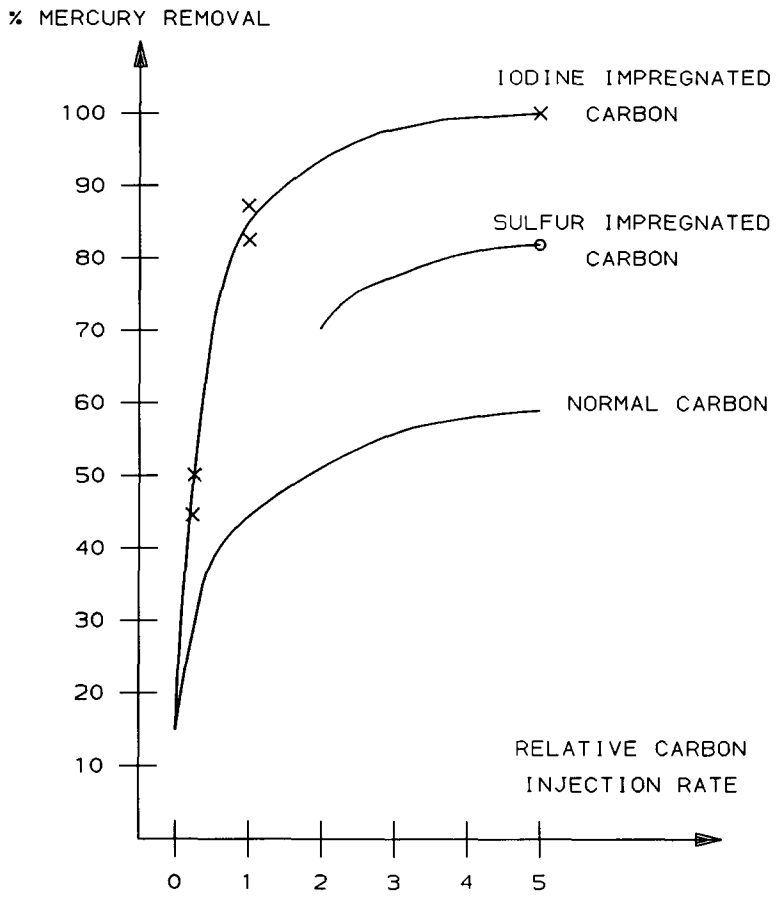

Fig. 6. Influence of active carbon type on mercury removal.

content and the percentage of oxidized mercury is shown in Fig. 7. It seems that even at low coal chlorine contents, a significant amount of oxidized mercury is present.

For eastern US coals, therefore, a dry scrubbing system can achieve high removal efficiency of mercury without any additional measures. For eastern low chlorine coal, the overall mercury removal efficiency can be enhanced by injection of normal activated carbon, which will be a relatively inexpensive method of achieving high mercury removal. For western coals with low chlorine content, the dry scrubbing system will be able to achieve a moderate mercury removal by injection of normal activated carbon, but for higher mercury removal, the more expensive iodine impregnated or sulfur impregnated carbon will have to be used.

The research work performed by Joy/Niro has only scratched the surface of mercury removal processes for coal-fired power stations. Further research work is

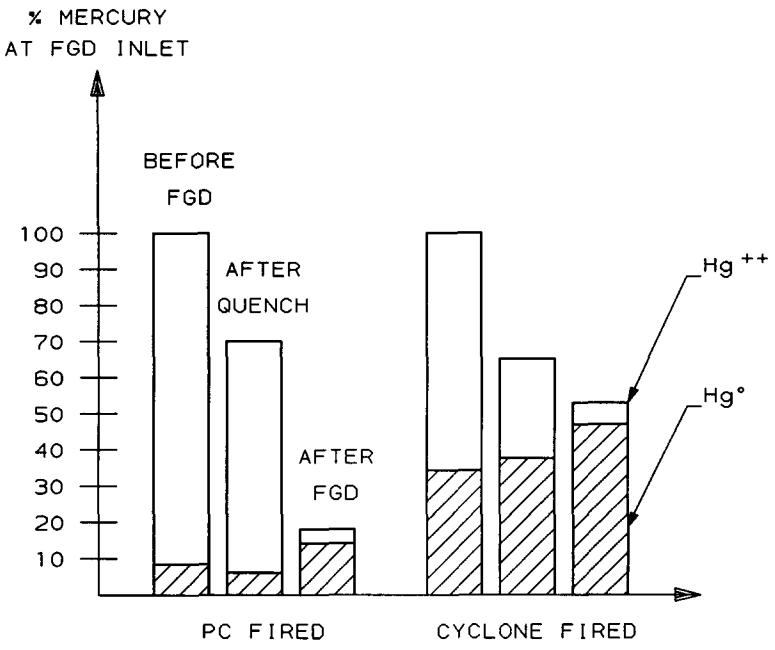

Fig. 7. Mercury removal in wet scrubber [19].

required to optimize the carbon type, as well as the costs aspects of the above-mentioned processes.

3. Control of other air toxics

Control of air toxic trace elements that are either not volatilized during coal combustion or are volatilized during combustion and later condense and coalesce in the form of fine particulates will generally be controlled by the removal efficiency of the dust collector. When a baghouse, either reverse air or pulse jet, is used, removal efficiencies of most trace elements in these categories are greater than 99%. These removals can further be improved through the use of ultrahigh efficiency fabrics. Membrane-coated fabric filter bags can reduce total emissions significantly and correspondingly reduce emissions of trace toxics.

One example of the removal efficiency of air toxics can be found in the Trace Metal Mass Balance Study made at the Studstrup Power Station. At Studstrup in Denmark, two 350 MWe units are equipped with spray dryer absorbers followed by pulse-jet fabric filters. A mixture of eastern US coal (West Virginia) and coal from Columbia, South America, is burned. The sulfur content in the mixture varies from 1.6% to 2.5%. The operating experience and SO_2 removal efficiency have been covered in a number of other papers [20–22].

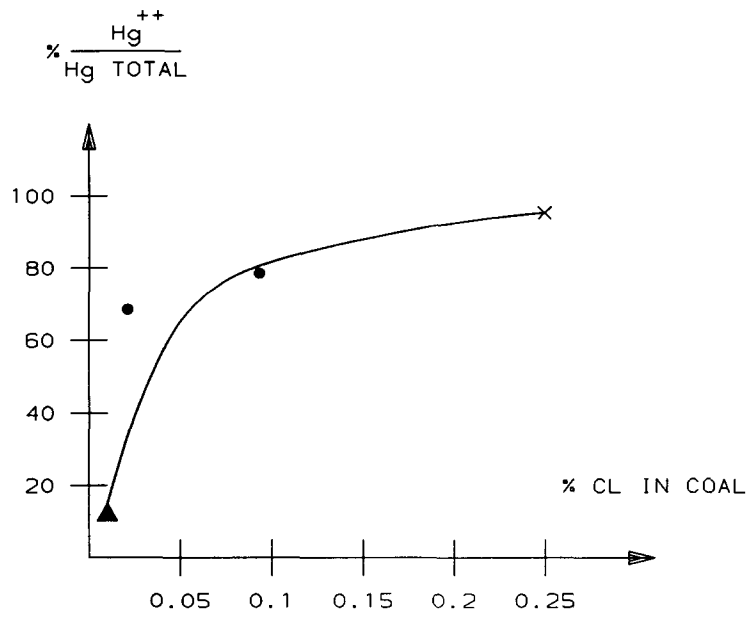

Fig. 8. Conversion of Hg^0 to Hg^{2+} as function of coal chlorine content.

Table 6
Studstrup trace metals emissions (g/h)

Metal	In (coal, etc.)	Emission	% Reduction
As	209.5	0.30	99.9
Cd	15.9	0.17	98.9
Cr	1514.0	4.00	99.7
Pb	344.0	8.00	97.7
Se	415.0	1.00	99.8
B	4499.0	44.00	99.0
Ni	975.0	8.00	99.2

In 1990, the Danish Power Station Engineering Company, ELSAM Projekt, performed a mass balance study, including trace metals at the Studstrup Power Station, Unit 3. During the mass balance study, Columbian coal with a sulfur content of 0.8% was burned.

For most of the trace metals, the mass balance could be closed within $\pm$ 20%. For arsenic and mercury, a larger deviation between ingoing and outgoing quantities was found. Table 6 shows the total trace metal amount into the plant coming from the coal, process water and lime. The contribution from the incoming trace metals by the lime was not insignificant. Ten percent (10%) of the incoming lead, for example,

Table 7
Vapor phase trace metal removal

Plant	Selenium			Boron		
	Inlet	Outlet	% Removal	Inlet	Outlet	% Removal
Studstrup 350 MWe	335[a]	0.8	>99	3600	35	>99
SH-pilot	80–250	<0.15	>99	1400–2200	70–150	93–95
NSP-pilot	30–59	1.3–3.7	>91	4300–8300	110–150	97–99
Wet[b] Scrubber			40–60			70–80

[a] Concentration in $\mu g/N\,m^3$.
[b] From an IEA study [23].

originated from the lime. The emissions in Table 6 are the combined gas phase and solid phase quantities.

As can be seen from the table, a high trace metal removal is achieved. This is also the case for boron and selenium, which were found in vapor phase.

In Table 7, the vapor phase removal efficiency for selenium and boron achieved at the Studstrup plant is compared with similar measurements at the SH pilot plant and the NSP pilot plant. As can be seen, a good agreement exists, and removal efficiencies of the order of $\gtrsim 90\%$ for both vapor phase metals are achieved. For comparison, wet scrubber data from an IEA study are included. For both selenium and boron, much lower removal efficiencies are achieved in a wet scrubbing system.

4. Revolatilization and leaching

By the enhanced SDA process for mercury removal, the mercury is transferred from the flue gas to the dry FGD product. However, if the captured mercury, later on during disposal of the dry product, is released again to the atmosphere, the mercury emission problem has not been solved. The concern for revolatilization of mercury has been addressed by Joy/Niro in several studies over the last three years [13,15]. The studies have concluded that no significant change in the mercury content of the dry product occurred, regardless of whether the product was disposed of in open air or contained in plastic bags. Thus, the results confirm that the mercury stays in the product both with and without active carbon injection.

Early studies on SDA product from coal combustion [28] from various plants confirmed that leaching of trace metals, especially mercury, from dry FGD products has not presented any environmental problems. Results of recently performed leaching studies have confirmed that the mercury, once captured, is stable and stays in the product, thus alleviating any concern for leaching of this toxic trace element.

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