

## Removal of Acid Gases in an Incinerator of Hospital Waste

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Abstract: At combustion of hospital waste numerous harmful pollutants were formed inclusive persistent PCDD/F (polychlorinated dibenzodioxines and dibenzofuranes) compounds. To eliminate these substances an existing incinerator consisting of the furnace heat exchanger with flue gas cleaning was used. This incinerator had a capacity of 250 kg of waste per hour. Elimination od acid gases was executed by dosing of solid sorbents (calcium hydrate and bicarbonate) at two points of the waste gases duct. Local temperatures of waste gases at these two points were different. Sorbents were dosed at different distances from the textile filter by which ash and sorbent were separated from the stream of waste gases. By this arrangement it was possible to alter the residence time of sorbents in waste gases. The effect of addition of bicarbonate on dechlorination and desulphurisation was also studied. The executed experiments indicated that at dosing of calcium hydroxide into waste gases at higher temperatures sulphur oxides reacted preferably to hydrogen chloride. The strong effect of HCl in waste gases on formation of PCDD/F was proved. For sufficient elimination of persistent compounds from waste gases it was necessary to add beside calcium hydrate also active carbon. Two types of sorbents based on carbon were tested, namely Norit and Chezacarb B. Both were macroporous. Chezacarb B was carbon black from the process of hydrogen production by partial oxidation. Efficiency of both sorbents for removal of PCDD/F was similar.

Key words: PCDD/F, adsorption, desulphurisation, dechlorination, active carbon.

#### 1. Introduction

In general wastes could be of different state (solid or liquid ) and could be dangerous to a different degree. Hospital waste is mostly solid and is always classified as dangerous. It is characterized by content of stable materials (glass, metallic and ceramic waste) and it also contains various waste drugs, PVC (plastics) and textile materials that are often infectious [1]. With regard to its properties it is usually stored in special combustible containers that are together with the waste charged into the incinerator. These are mostly rotary kilns or chambers with different type of grates. Flue gases contain beside acidic gases such as sulphur and nitrogen oxides also hydrogen chloride and persistent compounds inclusive PCDD/F. These pollutants are with regard to their global dimension regulated by emission limits. Small incinerators processing mostly medical wastes usually operate on basis of a dry process for elimination of dangerous pollutants. Dry sorbent is in this process dosed into the stream of waste gases and is then separated mostly by textile baffles in the filter.

Elimination of sulphur oxides from waste gases could be executed by many methods. Most usual is to use limestone [2] or calcium hydrate as a sorbent. If limestone is used for desulphurisation it is dosed directly into the kiln where it is at first calcinated and then it reacts with sulphur oxides. If calcium hydrate [3]

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is utilised for desulphurisation it is dosed into flue gases in the place where they have a temperature between 200 and 300 °C. There dehydratation followed with reaction of sulphur oxides takes place. Efficiency of this process is strongly dependent on temperature and humidity od waste gases which is the reason why in some casses water is sprayed into flue gases. Dechlorination is executed in a similar way as desulphurisation [4] with the exception of dry high temperature method. The formed salts of chlorides are unstable [5] at incineration temperatures .

In the dry desulphurisation method by calcium salts a significant increase of molar volume takes place which is the reason of formation of a sintered layer of sulphates on the surface. As the consequence of this effect the surface of particles is inactivated. Reactions of calcium oxide taking place at desulphurisation are shifted into the diffusion regime and for reaching of the required degree of desulphurisation in the given time excess of stoichiometric quantities of calcium salts is required. This effect applies also in dechlorination. Molar volumes for selected calcium and natrium salts are given in Table 1.

Molar volume of NaCl is smaller if bicarbonate is used for reaction of hydrogen chloride which is the reason why the surface reaction is controlled by kinetics of the reaction. Even more favourable conditions are when sodium carbonate is used. But different cases are with desulphurisation where the molar volume is larger in a similar way as with calcium salts. These are the reasons why at dechlorination it is more advantageous to use sodim carbonate, bicarbonate or trone. Bicarbonate is preferred as it is not forming hydrates. In spite of this fact, calcium salts are used in most cases for desuphurisation because of their low cost.

At combustion various fragments form from the fuel inclusive radicals. These fragments could contain beside other atoms also bonded chlorine. Hydrogen chloride in the oxidation atmosphere in presence of catalytically active particles of ash ( $Cu^{2+}$  or  $Mn^{4+}$ ) reacts with formation of chlorine [6]. This process known as the Deacon's reaction could be expressed in the form:

$$2 \times HCl + \frac{1}{2} \times O_2 \leftrightarrow H_2O + Cl_2 \qquad (1)$$

Chlorine is then the reactant and forms various persistent compounds that are appearing in the waste gas as harmful pollutants [9]. Reactions are taking place within the temperature range of 250 to 600 °C. For these reasons it is requested to reduce the content of hydrogen chloride in flue gases at temperatures above 600 °C. At these temperatures are dissociation pressures of chlorides very high with melting point of calcium chloride equal to 782 °C and of natrium chloride to 801 °C. These problems limit dehalogenisation of flue gas by standard sorbents at temperatures above 600 °C.

Emission limits of PCDD/F are at the level of 0.1 ng TEQ/m<sup>3</sup> at standard conditions. For these reasons there is a great affort to reduce further the content of hydrogen chloride and chlorinated compounds in flue gases. For further reduction of the formed persistent compounds physical adsorption or chemisorption are used [7, 8]. For reduction of emissions of persistent

Table 1 Density, molar mass and molar volume of selected salts.

Analyt	Density (kg/m <sup>3</sup> )	Mol. mass (g/mol)	Mol.volume (m <sup>3</sup> /mol)
CaO	3,320	56.08	16.89
CaCO <sub>3</sub>	2,710	100.09	36.93
CaCl <sub>2</sub>	2,174	110.99	51.05
CaSO <sub>4</sub>	2,930	136.16	46.47
Na <sub>2</sub> CO <sub>3</sub>	2,532	105.99	41.86
NaHCO <sub>3</sub>	2,217	84.01	37.88
NaCl	2,163	58.45	27.02
$Na_2SO_4$	2,664	142.05	53.32

compounds sorbents are used at temperatures lower than 250  $^{\circ}\mathrm{C}.$ 

### 2. Experimental Unit

Experiments were executed in the already existing incinerator plant of hospital waste. This plant had a chamber furnace where the waste was dosed in regular intervals. Capacity of the plant was 250 kg/hr. Temperature in the chambre furnace was kept at 900 up to 950 °C. Flue gases were pasing through the aftercombustion chamber, with the temperature of 1,100 up to 1,150 °C, to the steam boiler (steam generator). Temperature of waste gases at the outlet from the steam boiler was 200 to 220 °C. Into the stream of flue gases was 5-dosed calcium hydrate. Flue gases were passed through a 6-labyrint 1 which was 8 m long, into the heat exchanger where flue gases were cooled 130 to 135 °C. Finely crushed bicarbonate and sorbent based on 8-carbon were dosed into waste gases behind the cooler. Waste gases were then passed through the labyrint 2 which was 17 m long to the

10-textile filter(backhouse). There solid parts of flue gases were separated and purified waste gases were discharged into the environment. Volume of labyrint 1 was  $0.73 \text{ m}^3$  and volume of labyrint 2 was  $1.8 \text{ m}^3$ . Block diagram of the incineration plant is given in Fig.1.

## 3. Discussion of Obtained Results

# 3.1 Desulphurisation and Dechlorination by Calcium Hydroxide

Powdery calcium hydrate was dosed into the stream of waste gases behind the steam generator at temperature  $215 \pm 10$  °C. Flow rate of waste gases was 3,300 Nm<sup>3</sup>/hr. Their average composition is given in Table 2.

Flue gases were flowing through the labyrint 1 and cooler for hot water of volume 650 liters into the labyrint 2. Sorbent was then separated on the filtration baffle and waste gases were passed through the layer of catched sorbent. Total time of mutual contact of acid ghases with the sorbent dosed behind the steam generator was about 2.5 s.



Fig. 1 Block diagram of the incineration plant of dangerous hospital wastes.

		Concentration of analyte (mg/m <sup>3</sup> )					
	$SO_2$	HCl	NOx	$O_2$			
Average	534.1	121	782	10.3			
Uncertainity	$\pm 8.28$	$\pm 1.29$	$\pm 16.31$	$\pm 1.04$			

Table 2 Average composition of waste gases.

Concentration of  $SO_2$ ,  $NO_x$  and oxygen was measured by a continuous analyser and concentration of HCl was meassured by absorption into a solution with following argentometric titration. Measurements of gas concentration in flue gases were executed before dosing of sorbent and at the outlet from the textile filter, after separation of the sorbent. On basis of measured concentrations of acid gases conversion of the component *i* was calculated by use of the reaction:

$$X_{i} = \frac{(C_{i,0} - C_{i})}{C_{i,0}}$$
(2)

It was assumed that calcium hydrate reacted only with sulphur oxides and hydrogen chloride. Then theoretical consumption of hydtrate could have been calculated according to the equation:

$$G_{Ca(OH)_{2}}^{T} = 74.1 \times V \times \left( \frac{C_{SO_{2}}}{64.07} + \frac{C_{HCl}}{72.93} \right)$$
(3)

Theoretical consumption of calcium hydrate was for the given conditions 2,689 g/hr. The excess sorbent was the ratio of actually dosed calcium hydrate (G) and amount theoretically needed and calculated according to Eq. (3). Equation can be written in the form

$$\lambda = \left(^{\rm G}/_{\rm G^{\rm T}}\right)_{\rm Ca(OH)_2} \tag{4}$$

Dependence of conversion of sulphur oxide and hydrogen chloride on excess of dosed calcium hydroxide  $\lambda$  is given in Fig. 2.

It was obvious from these measurements that calcium hydroxide reacted with  $SO_2$  and HCl. Dependence of conversion of  $SO_2$  and HCl on the amount of added hydroxide into the waste gases was linear. Conversion of Ca(OH)<sub>2</sub> was determined by

analysis of ash taken from the textile filter and was 26 mass %. Content of chlorides in ash was 3.7 mass % and content of sulphur was 4.8 mass %. It was obvious from these results that calcium hydrate at sorption of acid gases adsorbed preferably oxides of sulphur as to those of hydrogen chloride.

In the next stage at the same conditions the point of dosing of the sorbent was changed. Calcium hydrate was dosed behind the heat exchanger of waste gases i.e. in front of labyrint 2. Temperature of waste gases at the dosing point was 130 °C. Dependence of concentratuon of SO<sub>2</sub> behind the textile filter was measured for various quantities of dosed calcium hydrate. Dependence of conversion of sulphur dioxide on excess of added calcium hydroxide for both dosing places is given in Fig. 3.

Dependence on conversion of sulphur dioxide on excess of calcium hydroxide was linear. Different slopes corresponded to changes of kinetic conditions, first of all of temperature and reaction time along the trajectory of motion of particles of calcium hydrate.

Residence time of sorbent in waste gases was 2.5 s at its dosing in front of labyrint 1. For the case of dosing of sorbent in front of labyrint 2 was the residence time about half of the first value, i.e. 1.3 s. By chemical analysis of ash from the textile filter concentration of calcium, chlorides and sulphates was determined. Conversion of Ca(OH)<sub>2</sub> was 20.2% and content of chlorides was 3 mass % and content of sulphates was 3.5 mass %. If results of both experiments were compared, it would have been possible to assume that dosing of calcium hydrate into waste gases with higher temperature (220 °C) would increase conversion of calcium hydrate and the ratio of Cl/S on reacted calcium sorbent would be reduced.



Fig. 2 Dependence of conversion of SO<sub>2</sub> and HCl on excess of calcium hydrate at dosing of hydrate into flue gases behind the steam boiler.



Fig. 3 The effect of dosing point of calcium hydrate into flue gases on dependence of conversion of SO<sub>2</sub> on excess of calcium hydroxide.

## 3.2 Desulphurisation and Dechlorination by Lime and Bicarbonate

Powdery calcium hydrate at the rate of 3.4 kg/hr was dosed into the stream of waste gases behind the steam generator and in front of labyrint 1. Flow rate of waste gases was 3,380 Nm<sup>3</sup>/hr. Their average composition measured in front of dosing of calcium hydrate is given in Table 3.

Also different quantities of bicarbonate were dosed behind the cooler of waste gases and in front of labyrint 2 while concentration of  $SO_2$  in waste gases was measured behind the textile filter. Dependence of concentration of  $SO_2$  on dosed quantity of bicarbonate is given in Fig. 4.

Experimental results demonstrate that concentration of  $SO_2$  decreased monotonously with increasing quantity of dosed bicarbonate. By analysis of the catched ash concentration of chlorides and sulphates was determined. Ash contained 3.6 mass % of sulphates and 4.5 mass % of chlorides. It was obvious that by addition of bicarbonates the quantity of eliminated hydrogen chloride had increased in comparison with desulphurisation only with calcium hydrate.



 Table 3
 Average composition of waste gases behind the steam generator.

Fig. 4 Dependence of concentration of SO<sub>2</sub> on dosed quantity of bicarbonate.



Fig. 5 Effect of excess of sorbent on conversion of SO<sub>2</sub>.

Comparison of conversion of  $SO_2$  in waste gases when as sorbent only calcium hydrate is added or if with calcium hydrate also bicarbonate is added is demonstrated in Fig. 5.

From these experiments results that addition of bicarbonate is increasing the amount of acid gases eliminated in flue gases. Addition of bicarbonate is changing the ratio of concentrations of chlorides and sulphates in ash which is in agreement with the assumed changes in molar volumes of natrium and calcium chlorides.

# 3.3 The Effect of HCl on PCDD/F and Their Division to Congeneres

At combustion (incineration) of compounds containing chemically bonded chlorine mostly hydrogen chloride forms but to a smaller extent also chlorinated radicals can form. Incineration takes place in an oxidation atmoshphere. Hydrogen chloride at such conditions could be oxidized chlorine that then reacts with organic radicals to various compounds inclusive persistent polychlorinated dibenzofuranes and dibenzodioxines. These compounds belong among those very closely regulated. Their emission limits are very low and they represent toxicity of individual congeneres.

The effect of content of hydrogen chloride on formation of PCDD/F in waste gases was measured. The content of hydrogen chloride was changed by addition of hypochlorite and crushed PET bottles into the hospital waste charged into the combustion chamber. No sorbent was added into waste gases. Samples of waste gases were taken in front of the textile filter where temperature of waste gases was 125 to 130 °C. Content of oxygen in waste gases was from 7.93 to 8.69% vol. Results of these measurements are given in Fig. 6.

It can be resulted from the made experiments that concentration of polychlorinated dioxines increases exponentially with content of chlorine in flue gases. This increase differs according to the type of congenere. The dependence of concentration of individual congeners of dioxines in flue gases is given in Fig. 7.



Fig. 6 Dependence of concentration of individual congeneres of dioxine on concentration of chlorides in flue gases.



Fig. 7 Dependence of concentration of individual kongeners of dioxines.

It is obvious from the obtained results that individual kongeners of dioxines are present at different levels where the largest presence is of hexachlor dibenzodioxines. Polychlorinated dibenzofuranes have also been studied. Their weighed sum gives information on total toxicity of flue gases. Comparison of presence of congeners of dioxines and furanes is given in Fig. 8.

It was obvious from the given experimental data that concentration of polychlorinated dibenzofuranes was nearly for an order of magnitude larger than concentration of polychlororinated dibenzodioxines. Most frequantly pentachlor dibenzofuranes were present, but dioxines were present most frequantly as hexa and heptachlor dioxines.

#### 3.4 The Effect of Calcium Hydrate

Calcium hydrate was dosed at the rate of 3.4 kg/hr behind the steam generator and observed was the decrease of concentration of hydrogen chloride and the change of concentration of dioxines. Concentration of hydrogen chloride was reduced from the original quantity of 558 mg/Nm<sup>3</sup> to 230 mg/Nm<sup>3</sup>. Decrease of concentrations of individual congeners of dioxin is given in Table 4.

From the presented experimental data, it is resulted that individual congeners reacted to addition of calcium sorbent in a different manner. Largest decrease of concentration was with tetrachlordibenzodioxines. With increasing number of chlorine atoms the decrease was slowly decreasing. This decrease was expressed as conversion. In Fig. 9 is given conversion of individul congeners of dioxines which is due to addition of calcium hydrate into the stream of waste gases in the point behind the steam generatore where the temperature is 220 °C.

The average decrease of concentration of all dioxines was 51.6%. It was assumed that at 220 °C dioxines already did not form. For these reasons it was obvious that calcium hydrate reduced concentration of hydrogen chloride in waste gases and also acted as a sorbent. This was the reason why also concentration of



Concentration of persistent compounds in flue gases with content of chlorine equal to 458 mg/Nm<sup>3</sup>. Fig. 8

Ca(OH) <sub>2</sub>	Chlorides	TeDD	PeDD	HexDD	HepDD	ODD	Total PCDD	
kg/hod	mg/Nm <sup>3</sup>	pg/Nm <sup>3</sup>						

Table 4 Change of concentration of dioxines and chlorides in stack gases which is due to addirtion of calcium hydrate.

Ca(OH) <sub>2</sub>	Chlorides	TeDD	PeDD	HexDD	HepDD	ODD	Total PCDD
kg/hod	mg/Nm <sup>3</sup>	pg/Nm <sup>3</sup>					
3.4	230	51	86	94	85	87	403
0	558	133	208	206	133	152	832



Fig. 9 The effect of calcium sorbent on reduction of concentration of kongeneres.

individual congeners was reduced. It was positive that the largest decrease of concentration was with tetrachlor dioxines, as this isomer had the largest impact at calculation of total toxicity of all polychlorinated dibenzodioxines.

#### 3.5 The Effect of Active Carbon on PCDD/F

It was not possible to comply with the emission limits for PCDD/F only by addition of sorbent suitable for elimination of acid gases such as sulphur dioxide and hydrogen halogenes. With small and medium size pollution sources for reduction of exhalations of persistent compounds adsorption on active carbon was usually utilised. With regard to the fact that waste gases contained oxygen it was necessary to dose active carbon into the stream of flue gases at a safe temperature. Another aspect was temperature at which adsorption on the surface was possible at the acceptable level.

Active carbon was dosed into the stream of waste gases behind the cooler at 120 °C. Active carbon was transported by a double screw conveyer from the storage tank into the mill where at high rotation speed carbon was crushed to fine particles smaller than 5 µm. Particles were then pneumatically transported by air and spread by a nozzle into the stream of waste gases. At high turbulence ( $Re \ge 10^5$ ) waste gases were passed through the labyrint 2 into the textile filter where solid

ash particles together with sorbent were sepatrated.

Content of SO<sub>2</sub>, NO<sub>x</sub>, HCl, Oxygen, and PCDD/F in waste gases was measured. Flow rate of waste gases was  $3,850 \text{ Nm}^3/\text{hod}$ . Average composition of waste gases is given in Table 5.

Calcium hydrate at the rate of 3.5 kg/hr was dosed into the stream of flue gases. Temperature in the point of dosing was 230 °C. Composition of waste gases measured behind the textile filter after addition of calcium hydrate is given in Table 6.

From comparion of concentrations, it is resulted that by addition of calcium sorbent not only reduction of concetration of hydrogen chlorides and sulphur oxides took place but also toxic equivalent of polychlorinated dibenzodioxines and furanes was reduced. But their concentration was exceeding the limiting values for more than one order of magnitude.

One of sorbents dosed to waste gas based on carbon was Chezacarb B, that was produced in the process of production of hydrogen by partial oxidation of distillation residuals of crude oil. Sorbent at the rate of 280 g/hr was dosed and concentration of PCDD/F was measured. The resulting concentrations inclusive flow rates of waste gases are given in Table 7.

With regard to the fact that concetration of PCDD/F was exceeding the limiting value the dosed quantity of Chezacarb was increased to 390 g/hr. The results of these measurements are given in Table 8.

		Concentration of analyte						
	$SO_2$	HCl	NOx	O <sub>2</sub>	PCDD/F			
Dimension	mg/m <sup>3</sup>	mg/m <sup>3</sup>	mg/m <sup>3</sup>	% obj.	ng TEQ/Nm <sup>3</sup>			
Average	245.4	458	188	8.49	4.852			
Uncertainity	$\pm 6.43$	$\pm 4.16$	$\pm 11.4$	$\pm 1.24$	± 35% rel			

#### Table 5 Average composition of flue gases.

#### Table 6 Composition of waste gases after dosing of 3.5 kg/hr of calcium sorbent.

	Concentration of analyte						
	$SO_2$	HC1	NOx	O <sub>2</sub>	PCDD/F		
Dimension	mg/m <sup>3</sup>	mg/m <sup>3</sup>	mg/m <sup>3</sup>	% obj.	ng TEQ/Nm3		
Average	196.2	331	196	8.65	3.187		
Uncertainity	$\pm 6.22$	$\pm 3.1$	$\pm 11.4$	± 1.25	± 35% rel		

#### Table 7 Concentration of analytes and flow rates of waste gases.

		Flow rate of flue				
	$SO_2$	HCl	NOx	$O_2$	PCDD/F	gases
Dimension	mg/m <sup>3</sup>	mg/m <sup>3</sup>	mg/m <sup>3</sup>	% vol.	ng TEQ/Nm <sup>3</sup>	Nm <sup>3</sup> /hr
Average	201.2	304	190	8.45	0.487	3,710
Uncertainty	± 6.22	$\pm 3.1$	$\pm 11.4$	± 1.25	± 35% rel	

#### Table 8 Concentration of analyte and flow rates of waste gases at larger quantities of used chezacarb.

		Flow rate of flue				
	SO <sub>2</sub>	HCl	NOx	$O_2$	PCDD/F	gases
Dimension	mg/m <sup>3</sup>	mg/m <sup>3</sup>	mg/m <sup>3</sup>	% vol.	ng TEQ/Nm <sup>3</sup>	Nm <sup>3</sup> /hr
Average	179	303.1	171	9.2	0.079	4,100
Uncertainty	$\pm 6.2$	± 3.3	± 12.8	± 1.5	± 35% rel	





It was obvious from the presented results that Chezacarb was an efficient sorbent for reduction of concentration of persistent compounds in waste gases. Specific origin of this sorbent was a limiting factor for its use. For these reasons identical experiments were executed also with classic active carbon NORIT. All instrumentation was identical as with dosing of Chezacarbu. It was calculated on basis of the obtained experimental data how many mg of sorbent had to be dosed into the stream of flue gases to reach the required level of toxicity of flue gases. Comparison of both sorbents is given in the next Fig. 10.

It was obvious from the obtained results that both types of sorbents were comparable and that it was not possible to give preference to one type of sorbent on basis of its sorption properties. To satisfy the emission limits for persistent compounds more than 0.1 g of active carbon had to be dosed per 1 Nm<sup>3</sup> of waste gases with composition of about 3 to 4 ng TEQ/Nm<sup>3</sup>.

#### 4. Conclusions

For elimination of acid gases and persistent polychlorinated compounds forming in small and medium size incinerator plants it is advantegeous to use sorbents of calcium salts, bicarbonate and acitive carbon. For ecologic and economic control of an incinerator, it is necessary to execute a number of operating experiments with the aim to obtain relevant data needed for decision making on the process of waste processing by incineration while fulfilling the specified emission limits for the given process. A suitable arrangement is combination of a stream adsorber combined with a following textile filter for removal of solid emisions together with the used sorbent. A good experience was made with combination of dosing of different types of sorbents separately into various points of the waste gas duct.

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