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# **Disposal of Gaseous Ozone-Depleting Substances (ODS) by High-Temperature Incineration**

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## Introduction

To protect the earth's ozone layer and to fulfil the international obligations under the 1987 Montreal Protocol, Hong Kong has put into effect legislation that control the import, export and manufacture of ozone depleting substances (ODS)<sup>1</sup>. As a result, spent ODS that can no longer be recycled or re-used have to be destroyed. Table 1 shows the major types of ODS generated in Hong Kong.

Table 1 Types of ODS in Hong Kong

Common Name	CFC-11	CFC-12	Halon-1211	Halon-1301 CBrF <sub>3</sub> -58				
Chemical Formula	CF Cl <sub>3</sub>	CF <sub>2</sub> Cl <sub>2</sub>	CBrCIF <sub>2</sub>					
Boiling Point (° C)	23.7	-29.8	-4					
Vapor Pressure at 20°C (Psi)	12.9	63.2	52.9 189					
Application	Used as	refrigerant	Used in fire extinguisher					

## **Overview of Incineration Technology for ODS**

There are several commercial ODS destruction technologies approved by the United Nations Environment Program (UNEP), such as

- liquid injection incinerator
- rotary kiln incinerator
- reactor cracking
- gaseous/fume oxidation
- cement kiln
- municipal solid waste incinerator (for foams only)

The primary overall products of thermal oxidation of ODS are carbon dioxide, water, hydrogen chloride, and hydrogen fluoride, and, in addition, hydrogen bromide (and/or bromine). By-products

<sup>&</sup>lt;sup>1</sup>Environmental Protection Department. *Environment Hong Kong 1998*, Government of the Hong Kong Special Administrative Region, 1998.

may also be produced as a result of incomplete combustion; these include carbon monoxide, carbon, hydrocarbons, organic acids, and any other waste constituents or their partially degraded products that escape thermal destruction in the incinerator. In well-designed and operated incinerators, these products of incomplete combustion (PICs) are emitted in minute amounts.

To enable the destruction of ODS be conducted in a reliable and environmentally acceptable manner, the incineration system must be equipped with the following features<sup>2</sup> -

- Proper construction materials throughout the system, particularly the combustion chamber and downstream gas scrubbing system, to withstand the corrosive acid gases formed in the destruction process;
- Scrubber design capacity for halogen acid gases removal;
- Entire system designed and operated to prevent or minimize the formation of PICs; and
- Ability to meet the destruction efficiency (DE) of greater or equal to 99.99% for the ODS.

Residence time and temperature have an effect on PIC formation and destruction. To minimize the formation of PICs, adequate residence time (generally one to two seconds), high temperatures, excess oxygen, and adequate turbulence or mixing of the compounds to be destroyed is required<sup>3</sup>. For the complete destruction of ODS, an adequate hydrogen source (methane or propane fuel gas, or water vapour) is required to promote the conversion of halogens to the acid gas form (HCl or HF) instead of free halogen gas (Cl<sub>2</sub> or F<sub>2</sub>). The incineration of bromine-containing halons may form HBr and Br<sub>2</sub>. There is a tendency to form Br<sub>2</sub>, which is difficult to absorb in the scrubber, even with an adequate hydrogen source. Previous researchers found that incinerating halogen-containing waste in conjunction with sulfur-containing waste produces beneficial results<sup>4</sup>. The SO<sub>2</sub> produced during the process apparently reduces the halogens that are present to the corresponding hydrogen halide, and no free halogens are detected. This reaction is particularly important during the combustion of brominated hydrocarbons such as halons. The introduction of a sulfur-bearing waste or an auxiliary fuel source containing sulfur (such as fuel oil) is in fact practised whenever brominated wastes<sup>5</sup> are incinerated.

<sup>2</sup>United Nations Environment Programme. Ad-hoc Technical Advisory Committee on ODS Destruction Technologies. May 1992.

<sup>3</sup>PEI Associates. Handbook : Pennit Writer<sup>••</sup> Guide to Test Burn Data (Hazardous Waste Incineration). EPA-625/6-86-012, U.S. Environmental Protection Agency, Cincinnati, Ohio, 1986.

<sup>4</sup>RES(TX) EDB Test Bum Program Emission Test Results: Volume I. Final Report. Prepared by Alliance Technologies. A-88-035. Bedford, Massachusetts, 1988.

<sup>5</sup>Whiting KJ, Irrgang GH. When is an Incinerator Not an Incinerator? Presented at 1990 Incineration Conference, San Diego, California, 14-18 May 1990.

### The Rotary Kiln Incinerator in Hong Kong

The Chemical Waste Treatment Centre (CWTC)<sup>6</sup> is an integrated facility to handle practically all types of chemical wastes generated in Hong Kong. It is equipped with a rotary kiln incinerator for the destruction of liquid and solid organic wastes. The system has been recently modified to treat gaseous ODS, and test bum has been performed to evaluate its performance in light of the UNEP and local requirements. This paper reports the results of the tests and highlights recommendations for future operation.

The CWTC incinerator consists of a horizontal rotary kiln and a vertical secondary combustion chamber (SCC) with a thermal capacity of about 9 MW each for the kiln and the SCC. Four lances at one end of the kiln allow pumpable waste streams to be injected. The opposite (discharge) end of the kiln is enclosed by a cylindrical breech, where ash falls through a bottom port into a submerged wet ash conveyor, and then into a 3m<sup>3</sup> tote box. The kiln is designed to operate at outlet gas temperatures from 700 °C to 1,000 °C.

Mounted on top of the discharge breech is the SCC where oxidation of the kiln gases is completed. The vertical chamber provides for a minimum gas residence time of 2 seconds, in an excess air environment. Two burners, each rated at 5.5 MW and capable of firing either waste fuel or No. 2 fuel oil, are mounted in the chamber to ensure that temperatures are adequate for destruction of organic compounds.

The combustion gases leaving the SCC enter the waste heat boiler. The unit is configured with two radiation passes and four connection passes, and is designed to lower the gas temperature from 1,100-1,200°C at the inlet, to approximately 345°C at the exit. During this cooling of the gases, heat is transferred to the boiler tubes to generate steam at a pressure of 17 bars. Slag built up is dislodged from the tubes by mechanical rappers. It is then collected and transferred via conveyors to 3 m<sup>3</sup> totes for proper disposal.

Following the waste heat boiler, the combustion gases pass into the gas cleaning system, which consist of a lime slurry spray dryer absorber (SDA), followed by a four compartment, pulse-jet fabric filter system. In the SDA, the gases are contacted by a finely atomized spray of hydrated lime and water. This spray serves two primary purposes : (1) it cools the flue gas from its entrance temperature of 345°C down to about 160-165°C at the exit; and (2) it removes the bulk of the acid gases from the gas stream, via the reaction with the lime. Typically, an excess of lime is used to ensure compliance with acid gas emissions criteria. Most of the dry powder formed in the spray dryer falls to the bottom and is removed in 3 m<sup>3</sup> totes. The remainder of the dried powder entrained within the gas stream then enters the fabric filter.

<sup>&</sup>lt;sup>6</sup>A hazardous waste facility of the Government of Hong Kong Special Administrative Region operated by Enviropace Limited through a design-build-operate contract.

In the fabric filter, the remainder of the acid gases and other contaminants (mainly metals) are removed as they pass through the filter cake on the fabric bags. The fabric bags are of woven fibreglass on a permeable membrane, which ensures that particulate and related emissions are minimized. Exiting from the fabric filter, the gases pass into the induced draft fan and then into the 76.2 m high stack. The induced draft fan is sized to overcome the incineration pressure drop while maintaining a slightly negative pressure at the kiln face plate, and therefore, throughout the entire system. The stack is equipped with both manual test ports and continuous emission monitoring (CEM) instruments. The incineration system is also equipped with automated waste feed cut-off system which is activated when the measured emissions exceed a pre-set percentage of the emission limits.

## **Process Modifications**

Though rotary kiln incineration is an approved technology, there is concern that the highly corrosive gases resulting from the combustion of CFCs/Halons may seriously shorten refractory life. Thus the primary goal of the test is to verify that the incinerator at the CWTC can achieve 99.99% destruction and removal efficiency while meeting all emission limits, and the secondary goal is to find out whether there is substantial corrosion that would prevent regular use of ODS as a waste stream. The following modifications are made to the incineration system -

- An ODS feeding system with automated flow controls, vaporiser, nitrogen purging and a system interlock (for emergency waste cut-off) is installed. ODS will be fed as a gas directly into the kiln for combustion.
- A 16 m<sup>2</sup> section at the roof of the radiation side of the waste heat boiler is covered with refractory lining for additional protection against corrosion.
- An additional lime slurry injection device is installed at the front plate of the kiln to render in-situ neutralization of acidic gases generated inside the kiln, and to raise the melting point of the solid waste mix to avoid premature melt down of the brick lining. Depending on the ODS feed rate, a dose rate of 0.3 MT to 2.4 MT of lime slurry (25% calcium oxide by weight) per hour is maintained throughout the combustion. In addition, bags of hydrated lime and glass are packed in boxes and are fed into the kiln through the automated drum feed system at a feed rate not less than 15 kg per hour to offer additional neutralization capacity.
- Waste oil with a high sulphur content ( around 1 to 2 %) is incinerated together with the ODS.
- A bare metal pipe is inserted into the SCC to evaluate the fluoride impact on the boiler tubes when burning ODS. The test section is made of boiler tube stock and water cooled from the inside.

#### **Test Procedures**

Four test runs were performed, with durations and ODS feed rates as in Table 2.

And the second	ODS	Test Duration (h)	Feed Rate (kg/h)				
Day 1	Halon-1301	8	30				
Day 2	Halon-1301	8	60				
Day 3	CFC-11	8	30				
Day 4	CFC-11	8	60				

Table 2 : Operation details for ODS test burn

Process parameters such as kiln and SCC temperature, stack gas flow and emissions were controlled as in Table 3 and closely monitored by the CEM system. The following parameters were measured continuously during the tests : carbon monoxide (CO), carbon dioxide (CO<sub>2</sub>), nitrogen oxides (NO<sub>x</sub>), sulfur dioxide (SO  $\frac{1}{2}$ , hydrogen chloride (HCl), oxygen (O  $\frac{1}{2}$  total hydrocarbons (THC) and opacity. The volumetric flow rate and temperature of the exhaust gas stream were measured and recorded by instruments not directly tied to the CEM system.

Table 3 : Operational Parameters for the Destruction of ODS

Parameter	Range of Values	Control Targets			
Kiln temperature SCC temperature	700 -1,200 °C 1,100 - 1,300 °C	1,000 °C 1,150 °C			
Gas residence time in combustion chamber Carbon content in ash	2-3 second none - 5 % (weight)	2.4 second 5 % (weight)			
O₂ in exhaust gas CO in exhaust gas	4 - 8 % (volume) 50 - 150 ppm (volume)	6 % (volume) 100 ppm (volume)			
Combustion efficiency [CO <sub>2</sub> /(CO <sub>2</sub> +CO)]	> 99.99 %	99.99 %			
Unburned hydrocarbons (as CH <sub>4</sub> )	5 - 50 ppm (volume)	25 ppm (volume)			
NO <sub>x</sub> emission	200 - 500 ppm (volume)	300 ppm (volume)			
HCI emission	0 - 38 ppm (volume)	22 ppm (volume)			
SO <sub>2</sub> emission	0 - 750 ppm (volume)	250 ppm (volume)			
Baghouse temperature	150 - 210 °C	170 °C			
Kiln draft	-0.14 mbar	-2.5 mbar			
Stack gas flow	27,000 - 38,000 m³/h	30,000 m <sup>3</sup> /h			

Grab sampling and analysis of stack emissions were also carried out for each test run .

Parameters to be determined are listed in Table 4. In addition, the following parameters were also measured: volumetric gas flow rate, moisture content,  $CO_2$  and  $O_2$ . Approved methods, such as those prescribed by the United States Environmental Protection Agency (USEPA), were used for both the sampling and analyses. These are listed below<sup>7</sup> -

- Method 1 : Sample and Velocity Traverses for Stationary Sources
- Method 2: Determination of Stack Gas Velocity and Volumetric Flow Rate (Type S Pitot Tube)
- Method 3 : Gas Analysis for Carbon Dioxide, Oxygen, Excess Air and Dry Molecular Weight
- Method 4 : Determination of Moisture Content in Stack Gases
- Method 5 : Determination of Particulate Emissions from Stationary Sources
- Method 8 : Determination of Sulfuric Acid Mist and Sulfur Dioxide Emissions from Stationary Sources
- Method 23 : Determination of Polychlorinated Dibenzo-p-dioxins Polychlorinated Dibenzofurans from Stationary Sources
- Method 29: Determination of Multi-metal Emissions from Stationary Sources

SW846-0030 : Protocol for the Collection and Analysis of Volatile PIC's Using a Volatile Organics Sampling Train (VOST)

Method 50 : Isokinetic HCI/Cl<sub>2</sub>, HF/F<sub>2</sub>, HBr Emission Sampling

There are six sampling points in the stack. For particulate/acid gas sampling, sulfuric acid sampling, metal sampling, and dioxins/furans sampling, three points were sampled in each of four ports. One of the remaining ports was used for CFC/Halon sampling. The sampling time per point and the total sampling time for each train are shown in Table 4.

Paramet	ter	Methods	Sampling Time Per Point (min)	Total Sampling Time (min)		
Stack Gas	Particulate, chlorine (as Cl <sub>2</sub> ), fluorine (as HF), hydrogen fluoride, hydrogen chloride, and hydrogen bromide	USEPA Method 5 and BIF Method 0050 (USEPA Method 50)	10	120		
11	Sulfuric acid mist and sulfur dioxide	USEPA Method 8	5	60		
4	Metals (antimony, arsenic, cadmiurn, chromium, copper, lead, mercury, and nickel) and total phosphorus (as P)	USEPA Method 29	10	180		
	Dioxins/Furans : PCDDs and PCDFs	USEPA Method 23	15	180		
20100	CFC-11/Halon-1301	SW846 Method 0030	15	180		
Feed	CFC-11/Halon-1301	Grab gas sample into Tedlar bags (Halon-1301) or Vial (for CFC-11), ODS analysed by GC-MS	5 (per sample)	9-10 samples per test run		

Table 4 : Protocols for Grab Sampling

To evaluate the fluoride attack on the boiler tubes during the tests, two identical mild steel tubes of known tube thickness were inserted into the roof of the SCC (see Figure 1) when Halon-1301 was bumt (60 kg/h). The first one was inserted when Halon-1301 was being incinerated, while the

<sup>7</sup>Title40 of the Code of Federal Regulation (CFR), Part 60, Appendix D.

other was inserted when normal wastes (i.e. without ODS feed as control) were fed. The durations of the two tubes inside the SCC were exactly the same.

## Results

The incinerator operated very smoothly with regard to stack emission control during the test burn. The kiln and SCC temperatures were stable and there was no major upset of the system (see Figures 2 to 6). No stack emission control limits were exceeded throughout the tests and hence waste feed cut-offs during the ODS destruction process were not experienced. Stack gas sampling results also indicated full compliance with emission control limits and no significant difference was observed between burning CFC-11 and Halon-1301, apart from higher HBr when Halon-1301 was fed at 60 kg/h (Table 5).

Parameter	Results (mg/m <sup>3</sup> , or and 12% $CO_2$ ))	Results (mg/m <sup>3</sup> , condition of dry at 0 $^{\circ}$ C, 1 atm and 12% CO <sub>z</sub> ))											
Constant Mary	Halons-1301 30 kg/h	Halons-1301 60 kg/h	CFC-11 30 kg/h	CFC-11 60 kg/h									
Particulate	0.5-1.4	0.2-0.6	0.2-0.6	1.0-1.5	75								
Chlorine and compounds (as $Cl_2$ )	< 2.9-3.2	< 3.8-3.9	< 2.7-3.2	< 3.4	100								
Fluorine and compounds (as HF)	< 0.3	< 0.4	< 0.3	< 0.4	25								
Acidity (as sulfuric acid)	10.8-11.1	24.9-36.0	17.6-18.0	5.5-12.6	100								
Sulfur dioxide	17.1-17.3	107.4-117.2	125.1-127.5	13.2-21.0	750								
Hydrochloric acid	< 3.8-4.4	9.5-12.9	5.7-9.3	< 4.4	38								
Total phosphorus (as P)	< 0.079-0.080	< 0.052-0.094	< 0.054-0.092	< 0.072-0.121	7.5								
Hydrogen fluoride	< 0.8-0.9	< 0.9-1.0	< 0.8-0.9	< 0.9	7.5								
Hydrogen bromide	< 0.7-0.9	2.7-7.5	< 0.8	< 0.9	7.5								
Toxic metal I (Hg, Cd & Sb)	< 0.030-0.423	< 0.024-0.043	< 0.019-0.025	< 0.156-0.456	3								
Toxic metal II (Pb, Cu, As, Ni & Cr)	< 0.317-0.321	< 0.236-0.377	< 0.205-0.326	< 0.221-0.402	10								
Total PCDDs and PCDFs	0.0019-0.0020	0.0123-0.0226	0.0250-0.0281	0.0193-0.0313	0.1 ng/m <sup>3</sup>								

Table 5 : Stack Gas Monitoring Results

The results of DE are shown in Table 6 and all are above 99.99%, the limit set out by UNEP (see above).

Table 6 : Results of Destruction Efficiency (DE) for burning Halon-1301 and CFC-11

Test Run	Actual Feed Rate Attained (kg/h)	Destruction Efficiency (DE) (%)
Halon-1301	22.8-30.6	99.995-99.999
Halon-1301	45.1-47.3	99.998-99.999
CFC-11	31.3-32.3	99.990-100.000
CFC-11	54.0-62.2	99.996-99.998

The bare metal pipes inserted into the SCC to test for corrosion gave an alarming indication. There was significant reduction in tube thickness in the one when Halon-1301 was incinerated (58.8mm o.d. versus the original 60.5mm) compared with the one with normal waste feed (thickness unchanged at 60.5mm). The cause of this excessive corrosion is not known but it may be due to a kiln temperature that is too low to break up all Halon-1301 where there is a lime spray to react with the hydrogen halides. Alternately the feed rate of 60 kg/h for Halon-1301 may simply be too high. A third possible reason is insufficient lime spray in the kiln .

## A Second Test Burn

A second test burn was conducted to investigate the reasons for the excessive rate of corrosion. The main concern is in the waste heat boiler because the kiln and the SCC are refractory lined and comparatively simple to repair. The test burn was conducted under the following controlled conditions -

- installation of 6 additional test pipes (see Figure 7) throughout the radiation side of the waste heat boiler;
- variation of Halon-1301 feedrate from 10 to 20 kg/h;
- variation of lime slurry injection rate from 0.5 to 2 m<sup>3</sup>/h; and
- variation of kiln temperature from 900-1150°C.

Table 7 gives the feedrates and the durations for the second test burn. Essentially the same operational parameters as in Table 3 are used. For the second test only Halon-1301 was used and emissions were monitored by the stack CEM. No grab sampling was arranged.

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	Halon 1301 feedrate (kg/h)	Lime slurry feedrate (m3/h)	Kiln temperature (degree C)	SCC temperature (degree C)	Duration(h)	Actual quantity incinerated (kg)			
Period 1	10	2	1100	1150	48	425			
Period 2	10	2	1050	1150	48	426			
Period 3	10	1	1100	1150	48	431			
Period 4	d 4 10 1		1050	1150	48	431			
Period 5	20	2	1100	1150	48	721			
Period 6	20	2	1050	1150	48	994			
Period 7	20	1000	1100	1150	48	973			
Period 8	20	1	1050	1150	48	981			
Period 9	20	1	1000	1150	48	1039			
Period 10	20	0.5	950-1000	1150	48	877			
Period 11	20	0.5	950-1000	1150	48	1062			

I able /. Flow rates for the second test bum

### **Results of the second test**

The outside diameters of the test pipes were measured by vernier callipers after every test period. Figure 8 shows the measurements after the first 4 test periods. Results show that under the operation conditions listed above, no noticeable corrosion is found. This is in sharp contradiction with the first test.

The explanation for the difference is quite simple. In the first test water at room temperature (25 degrees C) is used for the cooling of the test pipe. In the second test, blowdown water at 150 degrees C is circulated as cooling water through the test pipes. At the temperature of the first test, water vapor and acid gases in the flue condense to form a corrosive condensate. By elevating the temperature of the test pipes so that the dew points of the acidic conponents are exceeded, this type of corrosion is eliminated.

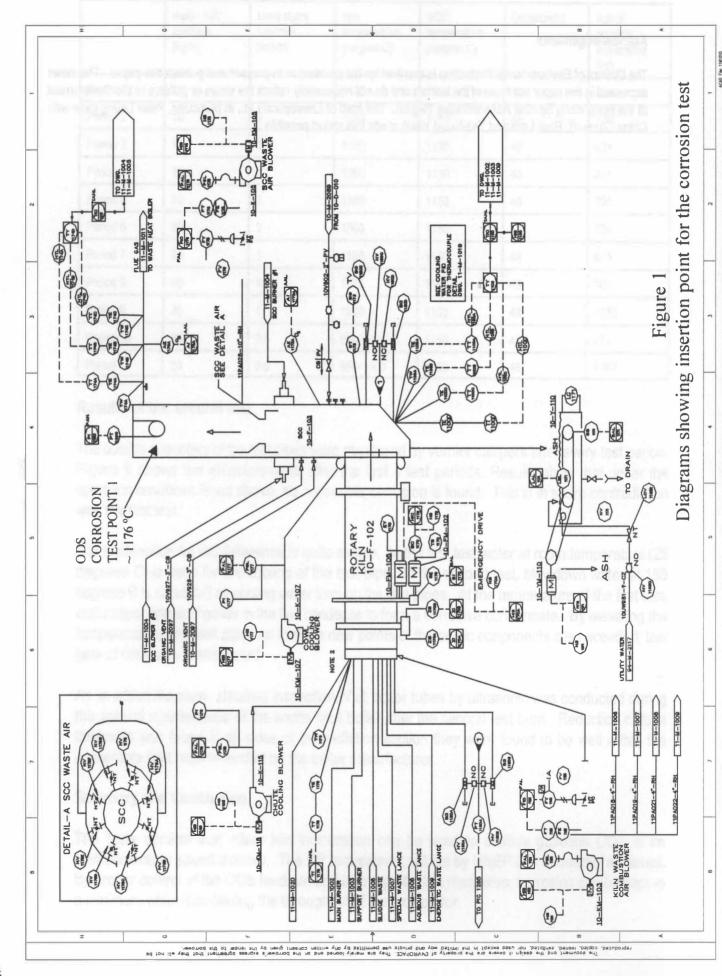
As an added measure, detailed inspection of all boiler tubes by ultrasonic was conducted during the annual maintenance of the waste heat boiler after the second test burn. Reduction of tube thickness was found in all sides of the radiation section they were found to be well within the acceptable limit recommended by the boiler manufacturer.

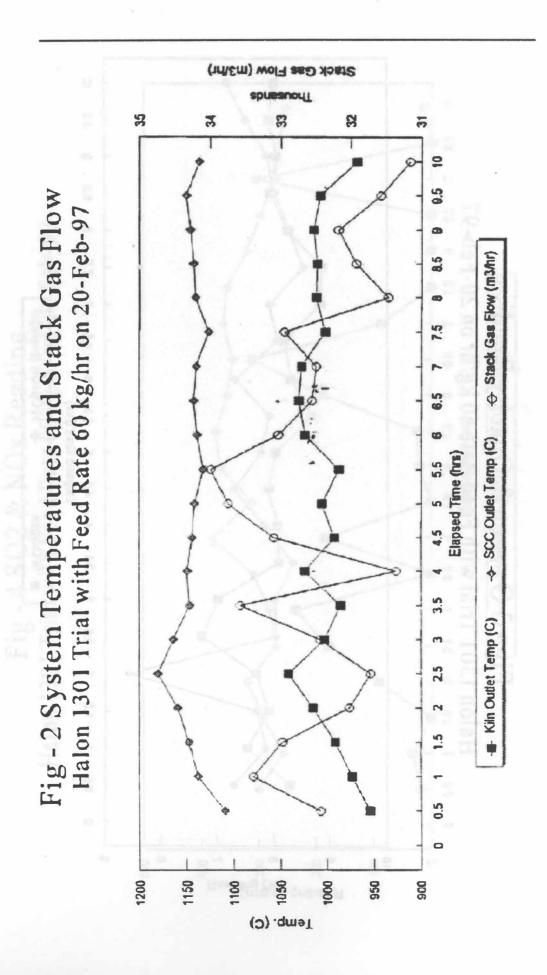
#### **Summary and Conclusion**

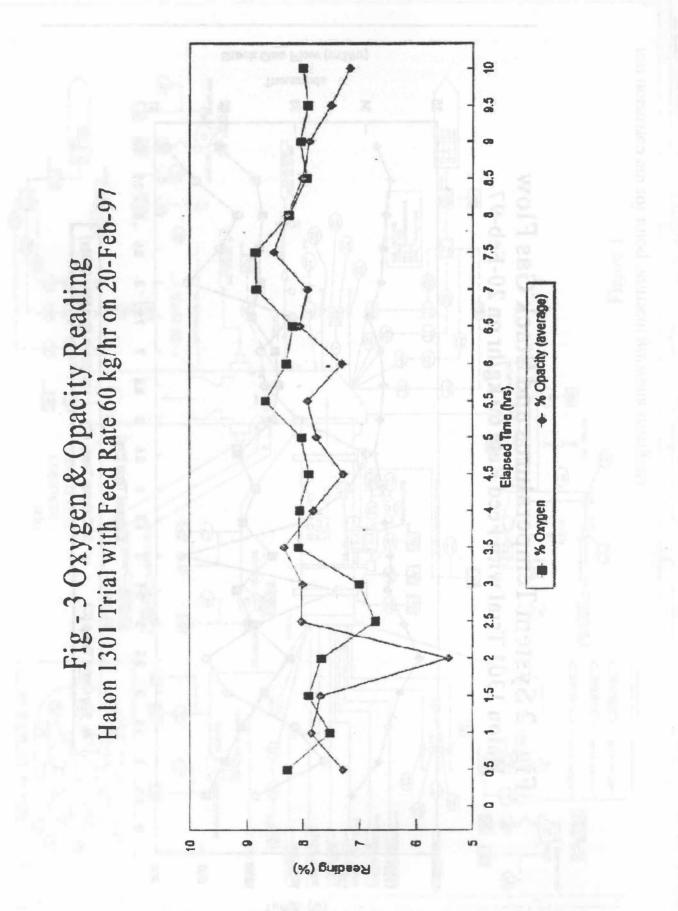
The tests confirm that rotary kiln incineration can be used to destroy gaseous ODS in an environmentally sound manner. The DE requirement set out by UNEP can be readily achieved. By proper control of the ODS feedrate and taking protective measures, corrosion can be kept to a minimum while maintaining the throughput of the incinerator.

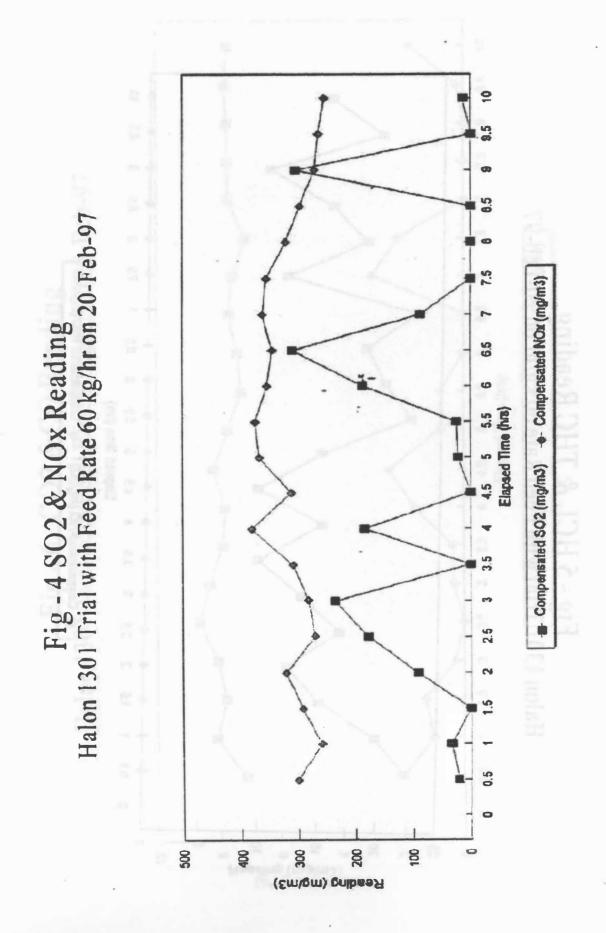
#### Acknowledgements

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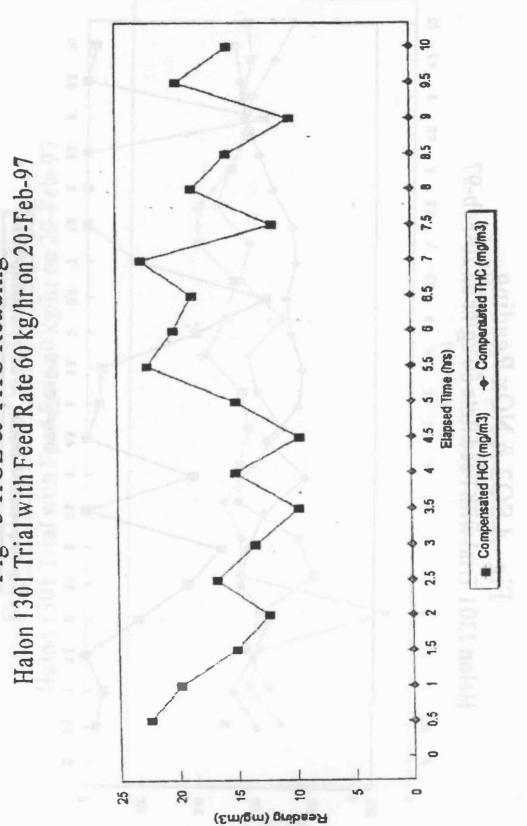
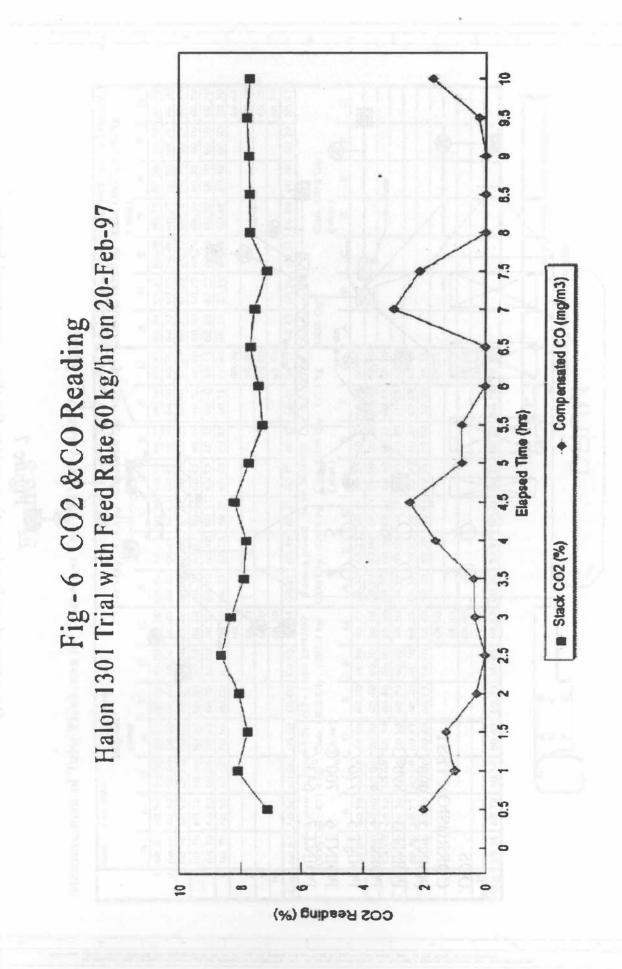


Fig - 5 HCL & THC Reading Halon 1301 Trial with Feed Rate 60 kg/hr on 20-Feb-97



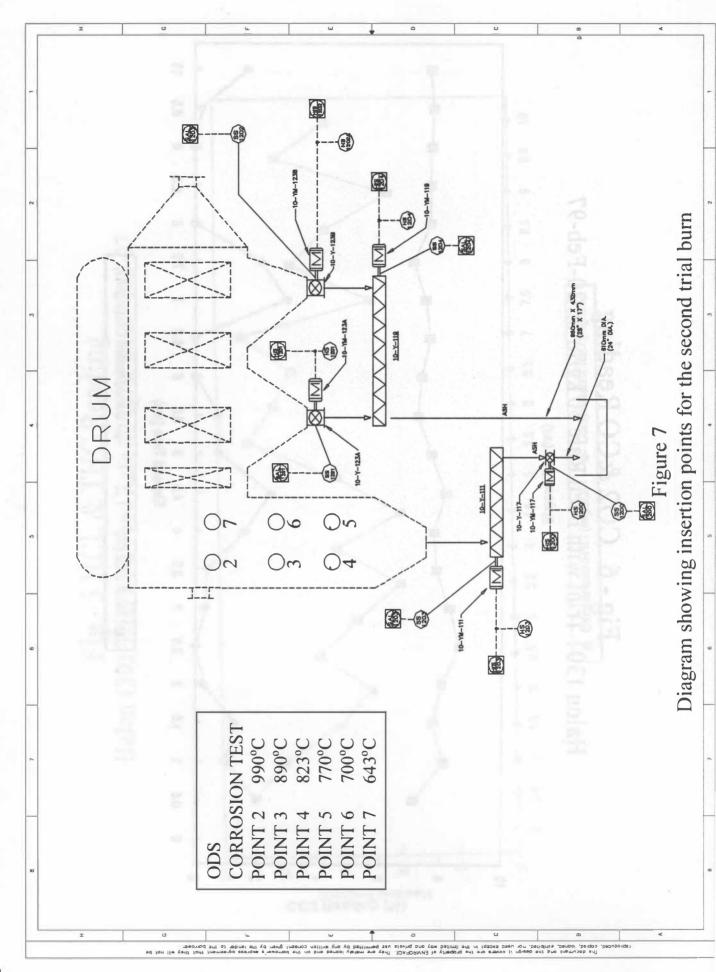
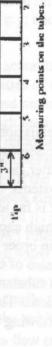


Figure 8

Measurement of Turke Thickness for Conosion Test of ODS - 11ALON 1301

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Testing period - 11.30 23/07/97 - 14:34 25/07/97	y: 12:	bn	0	60.35	60.45	60.50	60.43	60.40	60.25		Ser 19			01.00	& perk	1C+ : A)		B	63.25	60.00	60.00	40.96	59.95	07.68					59.97
11:163.1.	005 Qry : 125.3 kg		<	60.40	60.40	06.90	06.50	01.00	60.25		1	0		11.00	unes'l'	UDS UV : 430.6 kg		×	60.50	60.50	60.35	60.25	60.55	117 64			1	1	60,26
	10	0	0	00.00	60.40	60.40	60.30	60.40	101-10	66.30	60,40	60.50	60.30	60.44	160			5	60.40	60.40	60.40	60.50	121.04	111-05	<b>10,60</b>	55.90	69.40	601U	\$9.42 ·
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