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INVESTIGATION OF AN AEROSOL WITH PILOT UNITS INSTALLED ON SITE

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INTRODUCTION

The increasing concern over the pollution of air has in most countries led to stringent limitations on gaseous emissions. In the late 1960s a scrubber efficiency of 90 % was often considered to be most satisfactory. Nowadays an efficiency of 99+ % can be necessary to reach the prescribed emissions levels. It is very tempting then to install another scrubber of the same type and hope that this action will be sufficient. However, a great number of installations have failed to achieve anticipated efficiency levels in these situations. The cause of these failures is very often that fine particulates occur in the gas stream. These particulates can constitute the major part of the quantity emitted into the atmosphere after treatment in conventional control equipment. Conventional in this context means that the equipment is designed primarily for gas absorption. Fine particulates will pass through with low collection efficiency.

PROBLEM DESCRIPTION

Supra built a phosphoric acid concentrator in 1971. It was based on new and commercially untried technique with direct contact between the heat medium, which is flue gases, and the weak phosphoric acid. The hot flue gases are generated in a combustion chamber and accelerated uppwards through a venturi throat, where the acid is injected tangentially with three nozzles. The acid droplets move with the gas and the equilibrium between the acid and the surrounding gas is reached within a fraction of a second. When the droplets are separated in the cyclones the concentration of the acid has increased from about 30 % P_2O_5 to 54 % P_2O_5. The gases are treated in two TCA scrubbers placed in series.

A simplified diagram of the concentration plant can be seen in Fig. 1.

About 75 % of the F-content in the weak acid is evolved in this concentration step. This means that the inlet concentration of F to the first scrubber is 6000 mg/Nm^3 in the form of HF and SiF_4.

Fluosilicic acid was originally used as scrubbing liquid in both scrubbers. The concentration was 22 % in the first stage and 5 % in the second stage. Absorption capacity for the two scrubbers at those conditions can be estimated by using equilibrium data and well-known formulas for absorption efficiency. Such a calculation shows that the outlet concentration of F in the gas ought to be in the region of 10 mg/Nm^3 with two ball-beds in each tower.

Shortly after the start-up of the unit it was found that the emission of F was extremely high, about 1000 mg/Nm^3. By operating the two scrubbers at a lower concentration of fluosilicic acid it was possible to reduce the emission slightly. In order to improve the gas absorption further the second scrubber was modified so that sea water could be used on an once-through basis. The improvement in efficiency was small and not even after installation of spray nozzles in the gas duct after the second scrubber could the F-content in the gas be reduced below 300-500 mg/Nm^3.

The vendor of the pollution control equipment was consulted immediately after the discovery of the high emission level. It was suggested that the cause of the problem was channelling resulting in poor contact between gas and liquid. On the recommendation of the vendor the two towers were therefore lengthened by 2.5 meters without any improvement in efficiency could be noticed. Unfortunately the scrubber company was too unfamiliar with the application and could not provide any valuable information at all.

APPROACH TO THE PROBLEM

Some experiments were carried out by us during the following years. It was still assumed that better absorption conditions could solve the emission problem. All results were disappointing, however. Despite this fact several companies were willing to offer conventional scrubbers and also guarantee a maximum outlet F-concentration of 5 mg/Nm^3. Afterwards it is easy to show that none of the control devices offered to us at that time could have reduced the emission to any greater extent. This fact can only be partly blamed on the scrubber companies. We did not specify the problem correctly and they did not suspect fine particles to play an important role.

We knew by certainty that the National Franchise Board would refuse to grant a permission for prolonged operation of the phosphoric acid concentration plant. There were therefore three alternatives:

1. To evaluate the quotations and select a vendor for additional control equipment.
2. To start a thorough investigation of our own.
3. To shut down the unit and replace it with a concentrator of vacuum type.

The choise fell on the second alternative.

The aim of the investigation was to give a complete definition of the problem and to present an acceptable solution. An application to the authorities for a respite time of one year was granted.

The investigation started with an unprejudiced analysis of measurements being made so far. As mentioned before all efforts had been concentrated on actions improving the mass transfer. It was soon found out that the experimentally obtained variations of the F-concentration in the outgoing gas with alterations made in terms of different liquid flow rates, pH-adjustments of the scrubbing liquid etc. did not agree with gas absorption theory. As phosphorous also could be detected in the gas after the two scrubbers and phosphoric acid mist is a well-documented aerosol it was concluded that particulate scrubbing would be of interest in this case.

TEST PROGRAM

A two-step program was set up consisting of one introductory part and one experimental part.

The methods for particle size determination were surveyed. The measuring methods are more difficult and younger than those used in grain size determination and also less accurate.

Two attempts were made to determine the size distribution without any sophisticated instruments. Glass plates covered with a thin layer of sublimated MgO were inserted into the gas duct and exposed for a few seconds. The craters in the porous layer were photographed in microscope. The dots on the photograph were measured and counted so as to be able to plot a size-frequency curve. Calibration revealed however, that the method is too unsensitive to detect particles smaller than 5 μm. The measured droplets originated from the drop catcher and were 10-100 μm in size.

Next Millipore filters were tested. These filters are often used in laboratory work for particulate and biological analysis of fluid samples by vacuum filtration. Filters made of Teflon and cellulose ester and with pore sizes ranging between 0.8 μm and 1.0 μm were placed in filter holders and gas was sucked through this device. The best results were obtained with the cellulose ester fiber filter which reduced the F- as well as the P-content in the gas by almost 97 %. The conclusion from these experiments was that most of the particles seemed to be in the size range of $0.8 \lesssim d \lesssim 5$ (μm). Based on presented data on particulate removal efficiency for TCA-scrubbers, this interval could be further narrowed to $0.8 \lesssim d \lesssim 2$ (μm).

The preliminary investigation thus confirmed the particulate hypothesis and indicated the size distribution which is of outmost importance for the selection of suitable control equipment.

Literature survey gave an outline of the general capabilities of scrubbers and the circumstances under which they will perform at various levels of efficiency on fine particulate collection. The understanding and analysis of any scrubber can be reached by determining which combination of unit mechanism and particle deposition phenomenon that is involved.

Following particle deposition phenomena can be noticed:

Inertial impaction
Interception
Brownian diffusion
Electrostatic flux force
Gravitational force.

Only the first four have to be considered for extremely small particles.

After having analysed the problem so far the search for appropriate pilot plants followed. It was easier than expected to find companies having test units suitable for this purpose. Four types of equipment were finally chosen:

- Packed column
- Plate scrubber
- Brink mist eliminator
- Venturi scrubber

Sketches of the equipment can be seen in Fig. 2 and 3.

CONTACTING POWER THEORY

In addition to the pilot plants it was necessary to find a method which could be used for interpretation of data. An useful empirical method for correlating scrubber efficiency as a function of the power dissipated per unit of gas flow rate is the so called "contacting power theory" presented by Semrau et. al. The contacting power is the total power derived from both the liquid and the gas stream. This is given by the product of volumetric flow rate and the change in pressure. The functional relationship is expressed by

$$N_t = \alpha \cdot P_t^{\gamma}$$

where N_t is the number of transfer units and P_t is the contacting power. The constants α and γ are related to aerosol particle size and size distribution. N_t is defined by

$$N_t = \ln \left(\frac{1}{1-\eta}\right)$$

where η denotes efficiency.

Rearrangement of these formulas gives:

$$\log \ln \left(\frac{1}{1-\eta}\right) = \log \alpha + \gamma \log P_t$$

A logarithmic plot of $\ln \left(\frac{1}{1-\eta}\right)$ vs. P_t will thus give a straight line. This equation is applicable for all types of inertial collection equipment. All data obtained from the pilot plant tests were correlated by this relationship.

PILOT TESTS

The test units were mounted on the top plane of the concentrating unit at the same level as the gas duct to the stack. Gas was led in a PVC hose to the test units. Sampling was isokinetic and the gas was bubbled through two bottles filled with caustic solution. The F-content was determined by an ion selective electrode.

Division of the content by the measured gas sample volume gave the concentration in mg/Nm^3. Gas flow, liquid flow, pressure drop and temperatures were also measured at regular intervals.

RESULTS

Packed column

The reasons for testing a packed column were on one hand to compare and verify the results previously obtained from the large-scale tests and on the other to see whether a passage through the column could lead to particle growth.

Particle collection in packed columns can be described in terms of gas flow through curved passages. The performance can be predicted as being mainly a function of packing diameter (d_c) and bed depth (z). In this case Tellerette packing with d_c = 1" was used and the bed depth was 1.2 meters. Typical data are given below.

Pressure drop		40 mm WC
Gas flow		550 m^3/hr
Liquid flow		1000 l/hr
F-conc.	inlet	400 mg/Nm^3
	outlet	275 mg/Nm^3
η_F		31 %

An average efficiency of about 35 % was reached more or less independently of liquid rate. Performance prediction with respect to particle collection given in the Scrubber Handbook indicates a cut diameter[x] of 1.5-2.0 μm for a packed column under these operational conditions. The efficiency points are plotted in Fig. 4.

This column was later installed before two of the other experimental apparatus in order to study a possible condensation effect. The total removal efficiency was not significantly improved however, probably due to unsufficient condensation to give any detectable results.

Not surprisingly, this test with the packed column demonstrated the low collection efficiency on fine particles and, furthermore, cooling of the gas did not affect the particle size and enhance the efficiency.

Plate scrubber

A prototype of a horizontal plate scrubber developed by a Swedish company was next tested. The plate consisted of some kind of a plastic filter and was fixed between flanges, thereby making it possible to change the filter and vary the coarseness. The openings in the plates were 140-200 μm and this restriction caused a pressure drop of 15-150 mm WC per plate.

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[x] cut diameter = the particle diameter for which collection efficiency is 0.5 (50 %).

Scrubbing liquid under fairly high pressure (0.8 MPa) was injected co-currently. By changing the type of filter as well as the number of them in series a set of results for various total pressure drops were achieved.

Typical data:			
	Pressure drop		50 - 450 mm WC
	Gas flow		500 - 600 m^3/h
	Liquid flow		900 l/hr
	F-conc.	inlet	400 - 800 mg/Nm^3
		outlet	40 - 350 mg/Nm^3
	η_F		50 - 92 %

Scrubber efficiency as a function of contacting power is shown in Fig. 4. The linear relationship indicates that inertial impaction is the dominating deposition phenomenon. Otherwise a straight line would not have been obtained. The fan capacity limited the maximum gas pressure drop to about 500 mm WC. Extrapolation of the line to the desirable 98-99 % efficiency level gave a hint of the necessary pressure drop, somewhere in the range of 800-1200 mm WC. The large energy expenditure to achieve high removal efficiency led us to test a Brink mist eliminator.

Brink mist eliminator

This consists basically of a vertical packed fiber bed retained between two screens. The fibers are made of glass or synthetic material depending on the environment. Due to the presence of F, polypropylene and teflon fibers were used in our case. The collection efficiency for the so called H-E series elements is extremely good even for particles smaller than 0.1 μm. This is possible due to the utilization of Brownian movement as well as inertial impaction as controlling mechanism. Brownian movement or Brownian diffusion is defined as the random back and forth movement of fine particles caused by their collision with gas molecules. Hence, the probability that the particles will collide with a fiber and be collected increases as particle size decreases.

The phenomenon is illustrated in Fig. 5.

However, insoluble solids in a gas stream can be disastrous for Brink elements as plugging can occur and require periodic repacking. As soot and other inert material are present in the gas from the concentrator we were aware of the plugging risk. It is very difficult however, to predict the lifetime of the filter without carrying out some runs in pilot plant. In order to protect the main filter (H-E) a Brink H-V type mist eliminator was installed upstream.

Typical results:				
	Pressure drop		H-E	220 mm WC
			H-V	225 mm WC
	Gas flow			500 m^3/hr
	F-conc.	inlet		670 mg/Nm^3
		outlet		9 mg/Nm^3
	η_F (H-V)			85 %
	η_F (H-E)			99 %

The pressure drop change over the apparatus was closely followed. The test unit had been in operation for about 200 hrs when the gas flow dropped to almost zero on account of soot accumulation in the H-V filter. An examination of the H-E filter showed that soot also had penetrated the H-V filter and blackened the inner surface of the cylindrical filter.

The efficiency obtained in relation to energy consumed in terms of pressure drops was excellent as is shown in Fig. 4. A rough estimation of the cost for repacking of full-scale filters indicated that this would be prohibitive, however. The hope was therefore set on a venturi scrubber.

Venturi scrubber

The venturi throat and the cyclone separator were placed in between two high pressure fans. The pressure drop across the venturi was adjusted by changing the diameter of the plate in the throat section. Liquid was injected by a spiral type nozzle. The overall performance of the scrubber was in good agreement with the expected one. The efficiency data have been correlated to power requirement in the usual manner in Fig. 4. The desired removal efficiency level of 98-99 % was reached at a pressure drop of about 1000 mm WC.

Typical operational data:

Pressure drop		460-1130 mm WC
Gas flow		500-1100 m^3/hr
Liquid-gas ratio		1 l/m^3
F-conc.	inlet	580-870 mg/Nm^3
	outlet	5 - 30 mg/Nm^3
η_F		95.0-99.3 %

ANALYSIS OF THE PERFORMANCE DATA

The data points from the various apparatus, plotted in Fig. 4, follow the typical relationship discussed above. Two parallel straight lines can be drawn for the plate scrubber and for the venturi scrubber, respectively. The distance between the two lines reflects the difference in energy utilization. This means that the plate scrubber is uneconomic in comparison with the venturi scrubber if only the energy cost is considered. The reason is the high pressure drop on the liquid side for the plate scrubber.

The second thing to note is that the point for the Brink H-V filter is falling on the straight line for the venturi scrubber if this line is extrapolated. This confirms that the H-V type mist eliminator is designed to utilize impaction as the controlling mechanism of particle collection.

The outstanding performance of the H-E type filter, however, reveals that some other mechanism than inertial impaction must be involved, in this case Brownian diffusion. Considering the difference in energy cost between the H-E filter and the venturi scrubber it is not difficult to understand the intensive research in the field aming at the utilization of the other mechanisms. One example of this effort is the recently presented CDS (Charged Droplet Scrubber) which is a hybrid combining the best characteristics of electrostatic precipitators and conventional wet scrubbers.

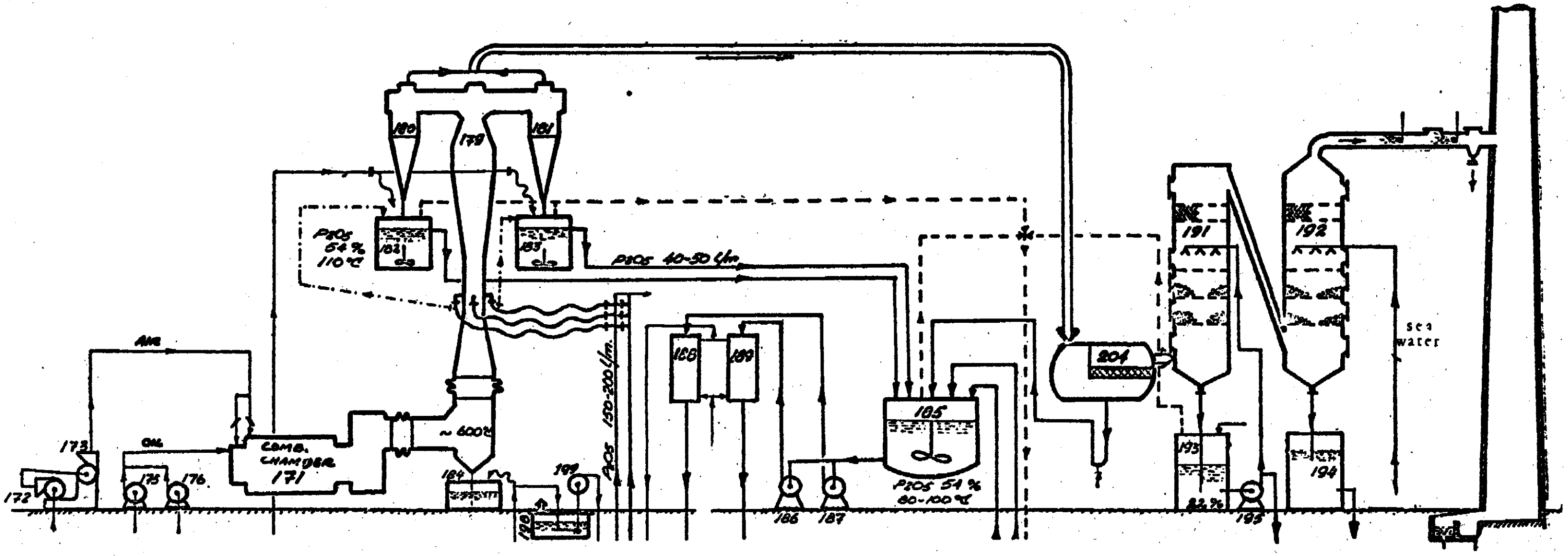
Fig. 6 shows correlation of scrubber efficiency, measured in transfer units, with contacting power for a series of aerosols, independent of type of scrubber.

DESIGN OF THE FULL-SCALE EQUIPMENT

On basis of this study a full-scale venturi scrubber was designed. Project engineering could be performed by ourselves thereby cutting the total cost for the project considerably. Equipment will be purchased from various vendors but the erection job will be done by us. There is of course some advantage in undivided responsibility. but the substantial cost for a turn-key installation makes this alternative of no interest in our case, as the risk for a failure has been minimized by this investigation.

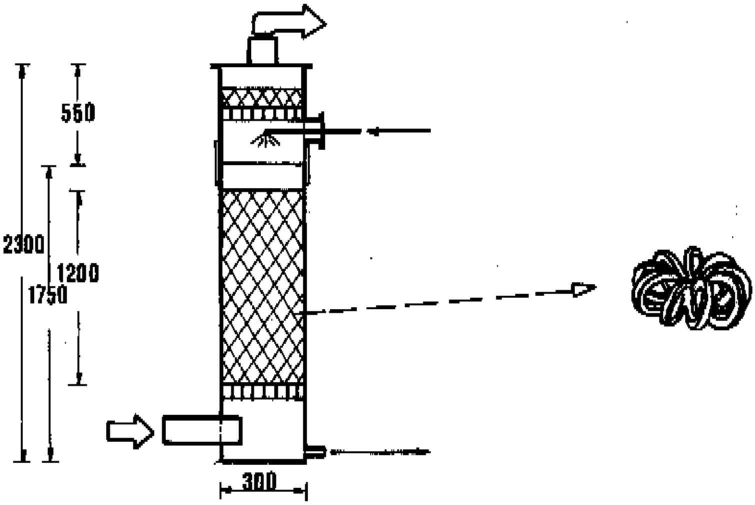
It took about 10 months to carry out this investigation and a total of 15 000 Skr ($ 3 500) was spent on equipment. We think this investment has been profitable and we have also learnt a lot of how to tackle emission problems.

FIGURE 1



phosphoric acid concentrator

FIGURE 2



packed column

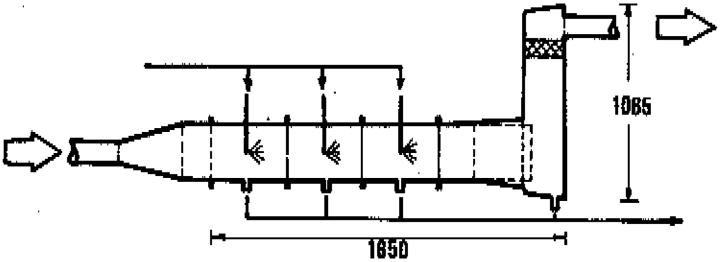
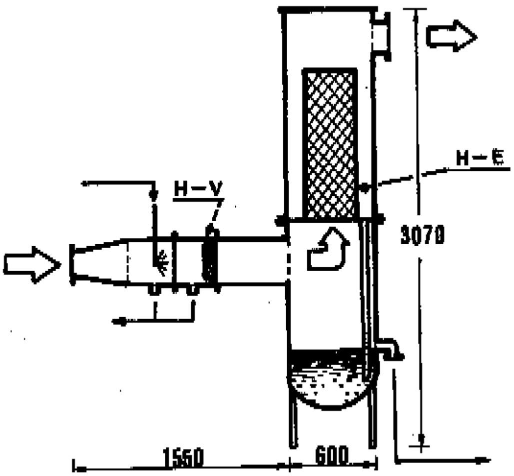
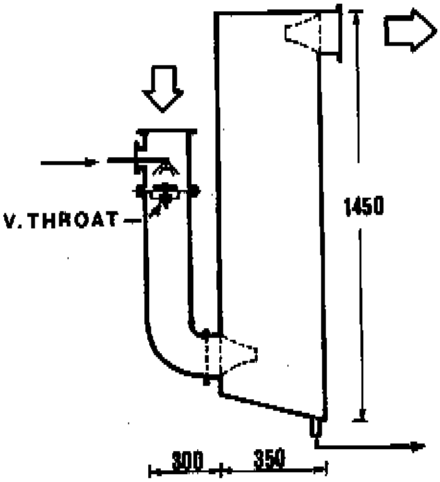


plate scrubber

FIGURE 3



Brink mist eliminator



venturi scrubber

SUPRA $\eta = f(P_t)$ FIGURE 4

2

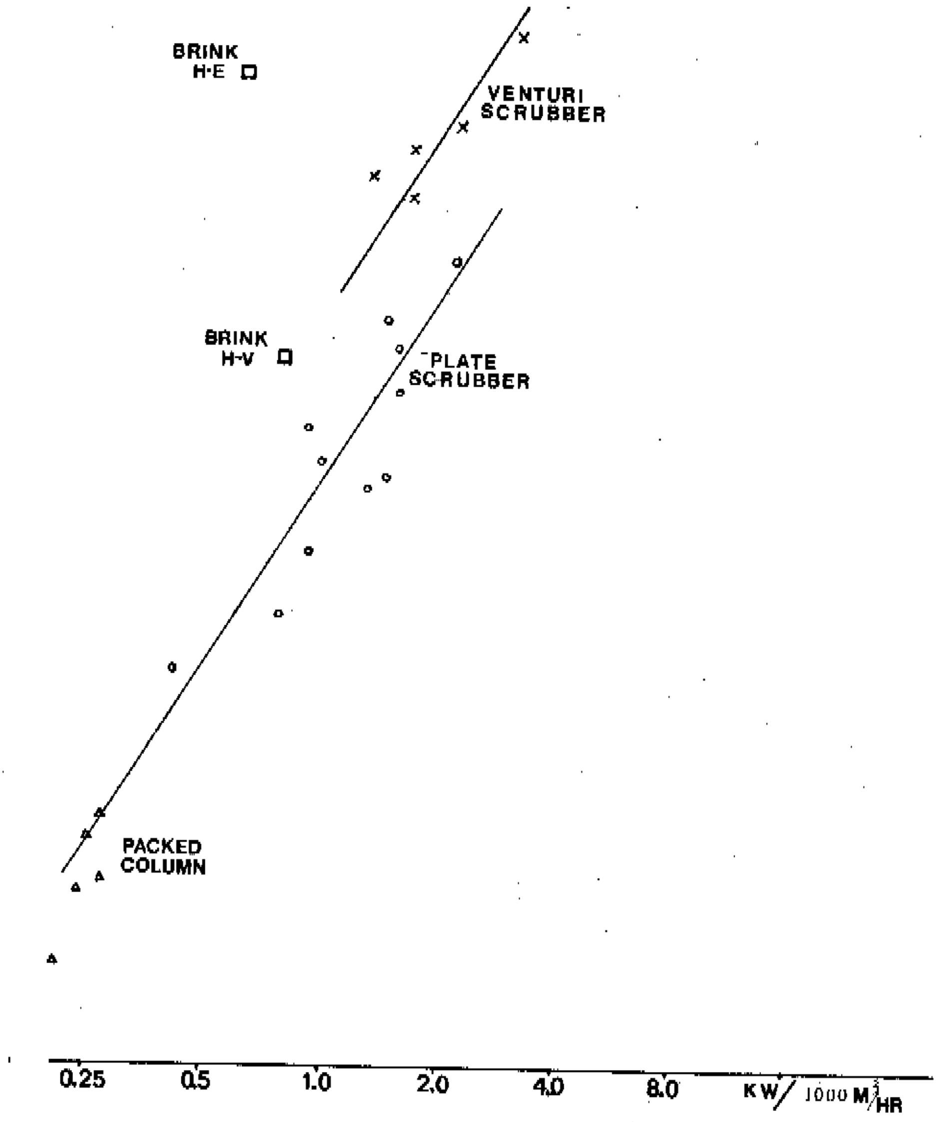


FIGURE 5

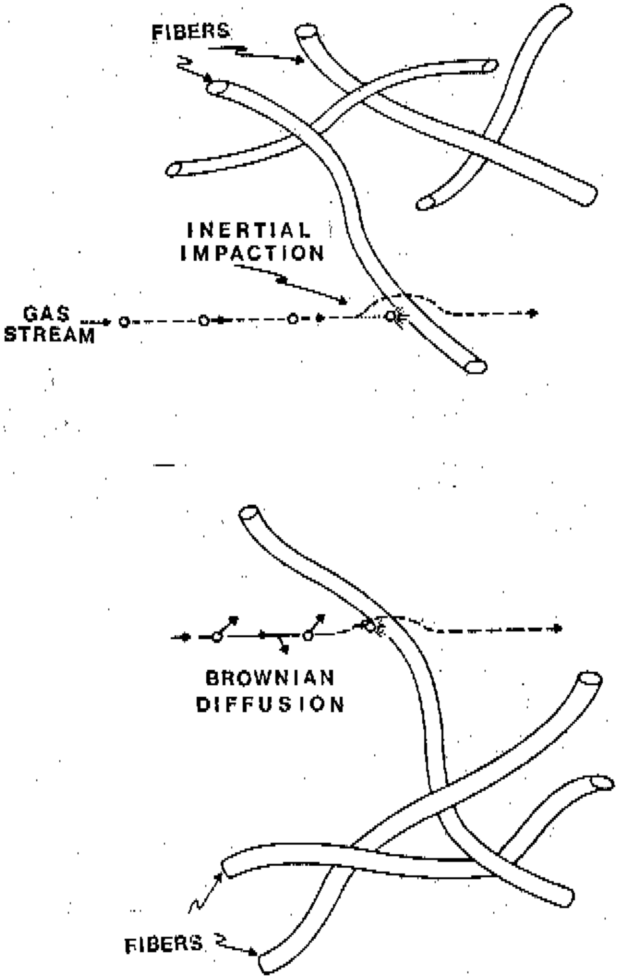
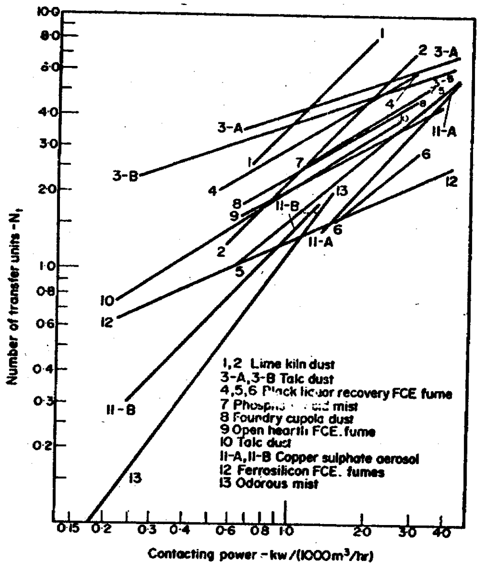

FIBERS
INERTIAL IMPACTION
GAS STREAM
BROWNIAN DIFFUSION
FIBERS

FIGURE 6



Performance curves for scrubbing of some aerosols

Curve	Aerosol	Scrubber	Correlation parameter	
			α	γ
	Lime kiln dust and fume (Kraft mud kiln)			
1.	Raw gas (lime dust and soda fume)	Venturi and cyclonic spray	1·47	1·05
2.	Prewashed gas (soda fume)	Venturi, pipe line and cyclonic spray	0·915	1·05
3.A.	Talc dust	Venturi	2·97	0·362
3.B.		Orifice and pipe line	2·70	0·362
4.	Black liquor recovery furnace fume	Venturi and cyclonic spray	1·75	0·620
	Cold scrubbing water (humid gases)			
5.	Hot fume solution for scrubbing (humid gases)	Venturi, pipeline and cyclonic spray	0·740	0·861
6.	Hot black liquor for scrubbing (dry gases)	Venturi evaporator	0·522	0·861
7.	Phosphoric acid mist	Venturi	1·33	0·647
8.	Foundry cupola dust	Venturi	1·35	0·621
9.	Open hearth steel furnace fume	Venturi	1·26	0·569
10.	Talc dust	Cyclone	1·16	0·655
11.A.	Copper sulphate	Solivore (A) with mechanical spray generator	0·390	1·14
11.B.		(B) with hydraulic nozzles	0·562	1·06
12.	Ferrosilicon furnace fume	Venturi and cyclonic spray	0·870	0·459
13.	Odorous mist	Venturi	0·363	1·41

From: W. Strauss
"Industrial Gas Cleaning" 2nd ed. Oxford 1975