Abstract
Air pollution is caused to a large extent by submicrometre liquid and solid particles called 'aerosols'. Separation of aerosols from residual gases is a major problem today. Well-known equipment like the venturi scrubber cannot handle submicrometre aerosols successfully. However, enlargement of the aerosols by condensed water vapour and subsequent separation by inertia forces in an air jet containing minute water droplets can lead to an acceptable process with moderate energy consumption. The generation of minute water droplets and their role in the separation of aerosols from gases is described in this paper.

Introduction
Most industrial plant procedures include cleaning of residual gases. Highly efficient systems like the venturi scrubber can successfully handle particles which are greater than 1µm. The cost and energy consumption of such systems if employed for the separation of submicrometre aerosols, in particular between 0.1 and 0.6µm, are very high and are unacceptable in most industrial applications. Enlargement of the aerosols by condensation and separation by impact is an elegant method to separate aerosols from residual gases of power plants, chemical plants and incinerators. The effect of the diameter of the water droplet and its relative velocity on the growth and separation of the aerosol is investigated here. A process of atomization which fulfils the conditions required for the separation of submicrometre aerosols and finally an apparatus for industrial application are described briefly.

Generation of aerosols
If reactions take place in the gaseous phase, they may lead to the generation of aerosols. The Walther process, developed by Krupp Koppers for flue gas desulphurization using ammonia, is considered here as an example /1/. This process was developed with the aim of establishing a desulphurization method with the following features:
- avoidance of wast-water and other waste products;
- production of the marketable nitrogenous fertilizer ammonium sulphate from the sulphur dioxide contained in the flue gases;
- reheating of the clean gases without external heating energy. Depending upon the partial pressures of the ammonia gas, sulphur dioxide gas and water vapour, the following reactions take place and generate aerosols from SO2, SO3, HCl and HF /2/.

(A) Aerosol generation by SO3:
\[
\begin{align*}
\text{NH}_3 + \text{SO}_2 + \text{H}_2\text{O} & \rightarrow \text{NH}_4 \cdot \text{HSO}_3 \\
2\text{NH}_3 + \text{SO}_2 + 2\text{H}_2\text{O} & \rightarrow (\text{NH}_4)_2 \cdot \text{SO}_3 \cdot \text{H}_2\text{O}
\end{align*}
\]

(B) Aerosols generated by SO3, HCl and HF:
\[
\begin{align*}
\text{SO}_3 + \text{H}_2\text{O} & \rightarrow \text{H}_2\text{SO}_4 \\
\text{H}_2\text{SO}_4 + 2\text{NH}_3 & \rightarrow (\text{NH}_4)_2 \cdot \text{SO}_4 \\
\text{H}_2\text{SO}_4 + \text{NH}_3 & \rightarrow \text{NH}_4\text{HSO}_4 \\
\text{HCl} + \text{NH}_3 & \rightarrow \text{NH}_4\text{Cl} \\
\text{HF} + \text{NH}_3 & \rightarrow \text{NH}_4\text{F}
\end{align*}
\]

These reactions take place in the gas phase and lead to ammonium salts which are hygroscopic and absorb water vapour from the environment to form a salt solution. The aerosols grow to a stable diameter depending upon the relative humidity in their vicinity. The generation of aerosols is a statistical process. Aerosols of various sizes with different salt masses may be generated. A separation process for salt aerosols with a diameter \(d_{50} \geq 0.05\mu m\) (corresponding to \(\pm 10^{-16}\)g salt mass) is described here.

Aerosol growth by condensation
The vapour pressure reduction at the surface of a water droplet of diameter \(d\) is, according to Kelvin /3/,
\[
p_p = \exp\left(\frac{4 \cdot \sigma \cdot \dot{M}_i}{d \cdot \Delta q \cdot \rho_L}\right)
\]

The vapour pressure reduction, \(p/p_s\), for pure water droplets is given in Fig.1 as a function of the droplet...
diameter $d$ for a water temperature of 50°C. Considering salt aerosols of ammonium sulphate with an initial pure salt diameter $d_s$ (in µm) and a salt mass $m$, the vapour pressure reduction on the surface of the aerosol /4/ (salt solution) is given by

$$\frac{p}{p_s} = \left(1 + \frac{6 \cdot \rho \cdot \sigma \cdot d \cdot \rho}{\pi \cdot d^2 \cdot \rho_{50}}\right)^{-1}$$

Combining eqns. (8) and (9), the vapour pressure on the aerosol surface is given by

$$\frac{p}{p_s} = \left(1 + \frac{6 \cdot \rho \cdot \sigma \cdot d \cdot \rho}{\pi \cdot d^2 \cdot \rho_{50}}\right)^{-1} \cdot \exp\left(\frac{4 \cdot \sigma \cdot \tilde{M}}{d \cdot R \cdot T \cdot \rho} \right)$$

This is indicated by the broken curves in Fig.1. The curves show the stable equilibrium diameter of the salt solution as a function of the environmental vapour pressure.

The salt aerosol with an initial salt mass of $10^{-16}$g will grow constantly as shown by the arrow A as long as the vapour pressure $p/ps$ is higher than 1.00224 (this corresponds to a pure water droplet of diameter 0.6µm). If the pure water droplets in the vicinity of the salt aerosol have a diameter of 2µm ($p/ps=1.001$), the equilibrium stable diameter of the salt aerosol is 0.26µm. For water droplets of diameter 6µm ($p/ps=1.00022$) the stable diameter of the salt aerosol is 0.24µm, as shown by the arrow B. The stable equilibrium diameter of the aerosol is 0.23µm for pure water droplets of diameter 26µm in the vicinity of the aerosol. The stable diameter for $p/ps=0.950$ (95% relative humidity) is 0.14µm. Aerosols with an initial salt mass of $10^{-15}$g ($d_{s_0}=0.11µm$) grow to a stable diameter of 0.3µm in an atmosphere with a 95% relative humidity, and to 0.6µm in a saturated atmosphere. It is evident from the curves in Fig.1 that the stable diameter is a very strong function of $p/ps$ (or the relative humidity). Considering $p/ps=1.00022$ (corresponding to pure water droplets of diameter 6µm), an aerosol with 10^{-16}g salt cannot be enlarged by condensation to diameters greater than 0.24µm.

Aerosols of about 0.3µm in diameter are very effective at scattering light and cause visibility effects similar to a summertime haze. Aerosols with an initial salt mass of about $10^{-16}$g ($d_{s_0}=0.05µm$) to $10^{-15}$g ($d_{s_0}=0.11µm$) leading to a stable salt solution (aerosol) diameter of about 0.25-0.6µm cause the major scrubbing problem. These aerosols cannot be enlarged by condensation unless the pure water droplets in their vicinity are equal to 0.6µm.

It is practically impossible to atomize water with a maximum droplet diameter of 0.6µm. A process to separate these aerosols by inertial forces is described below. The salt aerosols grow to 0.25µm within a very short time ($\approx 0.03s$) according to /5/:

$$t = \frac{\rho_{l} \cdot \tilde{M}_{g} \cdot (d^2 - d_{s0}^2)}{8 \cdot \delta \cdot \rho_{g} \cdot \tilde{M}_{l} \cdot \rho_{s} \cdot \left(\frac{p}{p_{s}} - 1\right)}$$

Aerosol separation by inertial forces

The aerosols enlarged to a diameter of over 0.25µm by condensation can be removed by impaction with high velocity water droplets. The removal efficiency is a function of the impaction parameter

$$\psi = \frac{\rho_{l} \cdot u_{R} \cdot d_{s0}^2}{18 \cdot \eta_{g} \cdot d_{L}}$$

and the Reynolds number

$$Re = \frac{u_{R} \cdot d_{L}}{v_{g}}$$

as shown in Fig.2 /6/.

The removal efficiency $\eta$ is 0.28 for a water droplet of diameter 6µm moving with velocity of 50ms^{-1} relative to the aerosol. This shows that, for acceptable inertial collection, water droplets of about 6µm are required. The relative velocity between the water droplet and the aerosol should be about 50ms^{-1}.

A process for the generation of minute droplets of diameter about 6µm moving with a high velocity relative to the aerosol particles is described here.

Generation of minute water droplets

Controlled atomization with a given maximum droplet size $d_L$ can be achieved by employing the low sonic velocity in a liquid/gas mixture. This process of atomization consumes less energy than do the pneumatic systems. The layout of an atomizing nozzle employing the low sonic velocity in a liquid/gas mixture is shown in Fig.3.

Liquid and gas, flowing with low velocities, are mixed together in a mixing chamber and accelerated to the sonic velocity of the two-phase mixture. The flow velocity at the outlet of the nozzle cannot exceed the
sonic velocity (critical mass flow rate). If the ambient pressure \( p_\infty \) is reduced, or the system pressure \( p_s \) is increased, a pressure jump \( \Delta p \) is achieved at the nozzle outlet \(/7/\).

The liquid/gas mixture is subjected to this pressure jump while flowing through the nozzle. The pressure in a liquid drop is higher than the ambient pressure by

\[
\Delta p = 4\pi \sigma / d_L^2
\]

where \( \sigma \) is the surface tension. It is postulated now that a drop with a diameter \( d_L \) is just stable if the pressure jump

\[
\Delta p_j = p_E - p_\infty = 2 \cdot \Delta p
\]

All the drops with diameters greater than \( d_L \) are unstable while flowing through the pressure jump and will be divided into smaller drops with diameters less than \( d_L \). Liquids can thus be atomized with a given (or required) maximum drop diameter. The maximum drop diameter is controlled by the pressure jump at the outlet of the nozzle.

**Design of the nozzle**

The pressure jump \( \Delta p_j \) at the end of the nozzle is given by

\[
\Delta p_j = 8 \sigma / d_L
\]

The pressure at the end of the nozzle is

\[
p_E = p_\infty + \Delta p_j
\]

The sonic (critical) velocity at the end of the nozzle is given by \(/8/\)

\[
\frac{a_c^2}{a_L^2} = \frac{\alpha}{\rho \rho_g (\rho_\infty / \rho_g - 1)} + \frac{1 - \alpha}{a_L^2} L \left[ 1 + \alpha \left( \frac{\rho_\infty}{\rho_L} - 1 \right) \right]
\]

\( \alpha \) represents the void fraction at the nozzle outlet. The properties of the liquid and gas phases such as \( \rho_\infty \) or \( \rho_g \) correspond to the pressure \( p_E \) at the outlet of the nozzle.

The intensity of the critical mass flow rate is

\[
\dot{m}_c = a_c \cdot \rho_{loc}
\]

with the local density

\[
\rho_{loc} = \alpha \rho_\infty + (1 - \alpha) \rho_L
\]

With \( f \) as the cross-sectional area at the outlet of the nozzle, the total mass flow rate is

\[
\dot{M} = \dot{m}_c \cdot f
\]

If \( x \) is the quality of the mixture, the liquid mass flow rate is

\[
\dot{M}_L = \dot{M}(1 - x)
\]

and the gas mass flow rate

\[
\dot{M}_G = \dot{M} \cdot x
\]

The void fraction \( \alpha \) is a function of the intensity of the mass flow rate \( \dot{m}_c \) and the quality of the mixture \( x \).

**Calculation procedure**

Any value of \( \alpha \) is assumed and \( a_c, \rho_{loc} \) and \( \dot{m}_c \) are calculated. \( \alpha \) and \( \dot{m}_c \) lead to the quality of the mixture \( x \) and to the required flow cross-section for given \( \dot{M}_L \).

**Energy consumption**

The frictional pressure loss in the nozzle is negligible compared with the acceleration pressure drop which is

\[
\Delta p_0 = \dot{m}_c \cdot a_c
\]

The liquid and gas pressure \( p_A \) required at the entrance of the nozzle is

\[
p_A = p_E + \Delta p_0
\]

The energy \( E \) required for the atomization is

\[
E = E_g + E_L
\]

The energy required for the compressed air is

\[
E_g = \dot{M}_g \frac{\kappa}{\kappa - 1} p_\infty \cdot \sqrt{1 - \left( \frac{p_A}{p_\infty} \right)^{(\kappa - 1)/\kappa}}
\]

and for the water pump

\[
E_L = \dot{V}_L (p_A - p_\infty)
\]

**Design of aerosol scrubber**

A layout of the scrubber is shown in Fig.5. Residual gas enters the scrubber at the point A. Pure water is atomized through the nozzle \( N_1 \) with a maximum droplet diameter of 6-26µm. Gas is saturated and cooled down to the wet bulb temperature. The aerosols present in the gas stream are enlarged to 0.24µm. Gas now enters the second
column from below and process water is atomized by the nozzle N₂. The enlarged aerosols are collected by impaction with water droplets. The water droplets are finally separated from the gas stream in the demister D.

This system has been employed in industrial plants in various components. A few typical applications are described here.

(1) Carbon black fumes are separated from the hot chimney gases originating in the gold recovery process of ‘Heraeus’ in Hanau. The gases are quenched and the aerosols are enlarged in the first stage. A second nozzle in series scrubs the enlarged aerosols. The fumes were visible at the chimney outlet before installation of the ‘CAS’ system. This plant has been working since 1980.

(2) A venturi scrubber was installed by Wiegand to clean the residual gases from an incinerator of the company Wacker-Chemie in Burghausen. The dust load at the venturi was about 200mg/m³ gas. Dust loads at the outlet of the venturi scrubber were 90mg/m³ gas without aerosol enlargement and 12mg/m³ gas with aerosol enlargement by the

‘CAS’-system. This plant has been in operation since 1985.

(3) Ammonia aerosols as described above were generated in a demonstration plant for the Walther process at the power plant in Mannheim. The dust load was about 250mg/m³. Aerosols of approximately 30wt.% less than 0.4µm were enlarged and separated in a plastic demister. The dust load was less than 10mg/m³ after scrubbing.

(4) This system is employed regularly in combination with the Waterloo scrubber. Many such plants are in operation in Canada, U.S.A. and elsewhere.

The energy consumption of this system is approximately 50% less than that of systems like the venturi scrubber [9] according to experiments performed at Hoechst AG in Frankfurt.

Nomenclature

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<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
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<tbody>
<tr>
<td>a</td>
<td>sonic velocity</td>
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<tr>
<td>d</td>
<td>diameter</td>
</tr>
<tr>
<td>f</td>
<td>cross-sectional area at nozzle outlet</td>
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<tr>
<td>i</td>
<td>number of ions per molecule of salt</td>
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<td>M</td>
<td>molecular weight</td>
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<td>m</td>
<td>mass</td>
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<td>ρ</td>
<td>density</td>
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<tr>
<td>σ</td>
<td>surface tension</td>
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Subscripts

A nozzle entrance  
c critical  
E nozzle outlet  
g gas  
j jump  
L liquid  
loc local  
R relative  
s saturation  
so solid

References


7 German Patent DE 2627880, C2, 1976.
