NEW TECHNOLOGIES FOR THE REMOVAL OF SUBMICRON PARTICLES IN INDUSTRIAL FLUE GASES


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1. Summary

Recent findings in toxicology showed the high toxicity of particles finer than 1 microns, that are currently not included in environmental regulations for industrial emissions. Nevertheless, social awareness of the possible correlations among particulate exposure and severe diseases in population living close to industrial areas is limiting the development of new industrial plants and, in some case, is driving population and governments to consider the shutdown of potentially harmful plants.

The scope of this work is to present a new technology, developed by Boldrocchi Srl and the Department of Chemical, Material and Production Engineering of the University of Naples “Federico II”, able to remove, with high efficiency, particles of submicron size. This technology, belonging to the family of wet electrostatic scrubbers, proved to be extremely useful in removing diesel particle matter included black carbon. It is envisaged that the new technology can overcome some of the limits of ESP and FF and can be suitably adopted in combustion plants so to allow safer operations for both workers and populations.

2. Introduction

Periodical alarms due to fine dust concentration levels in atmosphere, especially in urban or industrial areas, keep high the attention of public opinion on industrial plants emissions.

The capture of submicron aerosols from flue gases is a critical issue of environmental engineering. The actual collection efficiencies of the existing technologies are the minimum for particle diameters lying between 100 nm and 1 µm, in the so-called Greenfield gap. In this range both the mass and the diffusivity of the particles are too low to achieve efficient separations from the flowing gas (e.g. Seinfeld and Pandis, 1998).

Moreover, the issue of submicron particles capture is more and more topical. The evaluation of the environmental impact of an industrial plant, on the basis of dust residual mass flow from the stack, can’t ignore the analysis of residual dust particle size and its effects on public health. It is well known the dangerous effects of dust on human respiratory system is directly proportional to the number of particles and inversely proportional to dust size. So the same residual mass flow from the stack of an industrial plant has different impacts on air quality and population health if made of an high number of submicron grains in spite of few larger particles.

The capture of the particulate matter is usually carried out by fabric filters (FF) and Electrostatic Precipitators (ESP), which are the actual best available technologies.
However, these units are poorly able to capture particles of submicron or nanometres size. Moreover, an ESP is ineffective for particle resistivity higher than about $10^{11} \Omega \text{cm}$ or lower than about $10^8 \Omega \text{cm}$ and have a small tolerance towards water droplet contents entrained in the gas. Instead, a FF cannot be used if the water content in the flue gas can produce condense on the cake deposited on the bags.

We are now developing a new technology, known as wet electrostatic scrubber, WES, that consist in using an electrified spray of liquid (water usually) to remove the particles contained in a gas. The particles are sometimes pre-charged before entering the scrubbing chamber. Tests were reported in the open literature regarding particles and droplets charged with opposite or similar polarities, or with only the droplets or the particles charged. Although there are some industrial applications of WES and some international patents, general design and scale-up criteria for wet electrostatic scrubbers are not available at the moment.

The aim of the research Boldrocchi and Federico II University are developing is to evaluate the efficacy of WES technology in the capture of submicron particles carried in the residual emissions from large scale combustion plants, in the fields of energy or cement and steel production.

This paper describes WES processes by showing a simple model for particle capture and some application of the WES process to highlight the potentiality of WES technology and to assess preliminary design criteria.

### 3. Preliminary modelling of Wet Electrostatic Scrubbing

For the sake of simplicity this paper refers to an ideal case that considers a simplified description of the spray fluid dynamic and neglects the droplet electrostatic charging process. The goal of the model is to allow the assessment of preliminary design criteria for WES systems tracing the path for future investigations. The theory of the particles capture by droplet scavenging is mainly derived from studies on atmospheric aerosol scavenging during rainfalls (Seinfeld and Pandis, 1998).

According to this studies, the instantaneous scavenging rate for particles of diameter $d_p$, $\dot{n}(d_p)$, by a water spray of droplets with size distribution $N(D_g)$, numerical concentration $C_g$ and relative drop-gas velocity $U_g$ is given by:

$$
\dot{n}(d_p) = n(d_p, t) \int_{0}^{\infty} \left( \frac{D_g + d_p}{4} \right)^2 \cdot U_g \cdot E \cdot C_g \cdot N(D_g) \cdot dD_g = n(d_p, t) \cdot \Lambda(d_p)
$$

where $n(d_p, t)$ is the numerical particle concentration at the time $t$. The integral, named scavenging coefficient, $\Lambda(d_p)$, is defined over the entire range of droplet sizes and represents the inverse of the characteristic time for particle scavenging. The overall scavenging rate is modelled as a linear superposition of the contributes pertaining to each single scavenging drop. This is verified if the volumetric drop concentration is sufficiently low (Volume fraction of droplets, $\varepsilon <10\%$) to neglect droplet-droplet interactions, a well posed assumption for industrial wet scrubbers. The collision efficiency, $E$, resumes all the features of the droplet-particle interactions accounting for capture mechanisms related to inertial effects, $E_{In}$ (Slinn, 1983; Licht, 1988; Kim et al., 2001), hydrodynamic interaction, $E_{HI}$ (Slinn, 1983; Jung and Lee, 1998), Brownian diffusion, $E_D$ (Slinn, 1983; Jung and Lee, 1998), thermophoretic, $E_{TP}$, and
diffusiophoretic, $E_{DP}$, phenomena (Seinfeld and Pandis, 1998) and electrostatic interactions, $E_{Es}$ (Davenport and Peters, 1978). The numerical value of the overall collision efficiency $E$ is the sum of each collision efficiency contribution. A detailed description of the collision efficiencies is beyond the scope of this paper and can be found elsewhere (e.g. Seinfeld and Pandis, 1998). Anyway, it is worth noting the expression of the $E_{Es}$ given by Davenport and Peters (1978) model is:

$$E_{Es} = \frac{16Kc Cc qg qg}{3\pi \mu_{\text{gas}}} U_g \mu_{\text{gas}} d_p$$

In Eq. (2) $K_c$, $C_c$ and $\mu_{\text{gas}}$ are, respectively, the dielectric constant, and the particle Cunningham slip correction factor and the gas viscosity while $q_p$ and $q_g$ are the particle and the droplet charges. The capture of particles by electrostatic forces is higher for smaller particles and droplets moving at lower relative velocity. The droplet charge depends on the electrical charging system and it can be considered as a fraction of the so called Rayleigh limit charge $q_R = k q_R$, that is the highest electrical charge that can be present on a droplet of a given diameter, $D_g$, without making it unstable and eventually tearing it apart. The value of $q_R$ is given by:

$$q_R = e \sqrt{\frac{2\pi \sigma_D}{K_c e^2}}$$

where $e$ is the electron charge and $\sigma_w$ is the droplet surface tension. The charge on a particle mainly depends on its physical properties and its diameter as well as on its peculiar the triboelectric charging. Detailed studies on different aerosol types have been reported by Johnston (1987) and Marra and Coury (1999), which, by numerical fit of experiments, expressed $q_p$ as a function of $d_p$ and aerosol properties as:

$$|q_p| = Ad_p^a \cdot e$$

Recently also the charge distribution on motor vehicle exhaust particles have been studies by Matti-Maricq (2006). Figure 1 reports the values of the collision efficiencies in function of the particle diameter. It is evident that electrostatic interaction is the most relevant collision mechanism for particle diameter below 1 $\mu$m overwhelming all the other collisional contributions. Only for particle diameter higher then 1 $\mu$m the inertial impacts prevails.
The main goal of a WES is to achieve high collection efficiencies with low costs for equipment and process operations. The former is mainly proportional to the WES volume, i.e. to the required contact time and to the spray nozzles costs. The construction materials must be chosen to reduce electric dispersion with the ground and to assure safety operations. The operational costs are mainly proportional to the water consumption, which influences both the WES pressure drops and the cost for droplet charging.

In this study, the main process parameters for WES are considered to be the residence time and the water consumption, represented by a volumetric water fraction, $\varepsilon$.

In particular, this paper considers the ideal case of a gas flow ($P=100$ kPa, $T=25^\circ$C) containing a given concentration of coal dust modelled as spheres of the same size and uniformly distributed, which contacts an uniform water spray of identical monodisperse spherical droplets moving with the same relative velocity respect to the gas flow.

The particle population balance for a given $d_p$ is described by the equation:

$$\frac{d}{dt} n(d_p,t) = \dot{n}(d_p) = -n(d_p,t) \cdot \Lambda(d_p)$$  \hspace{1cm} (5)

whose solution, for a given initial concentration $n(d_p, 0)$ is:

$$n(d_p,t) = n(d_p,0) \cdot \exp[-\Lambda(d_p)t]$$  \hspace{1cm} (6)

Finally, the collection efficiency is:

$$\eta(d_p) = \frac{n(d_p,0) - n(d_p,t)}{n(d_p,0)} = 1 - \exp[-\Lambda(d_p)t]$$  \hspace{1cm} (7)

Figure 1 – Collision efficiencies for coal dust in contact with a single water drop ($D_g = 400 \, \mu m$, $U_g = 6 \, m/s$, $k=0.1$) in function of particle diameters.
In the following, Licht’s (1988) model is used to calculate $E_{in}$, Slinn’s (1983) equations are used for $E_D$ and $E_{HI}$ while Eq. (2) is used for $E_{E}$, and phoretic effects are neglected. The model is applied to reference conditions (Table 1) typical of industrial wet scrubbers. Coal dust particles, largely diffused in industrial facilities, are considered. In this case the parameters of eq.(4) are $A = 36.8$ and $B = 1.17$ (Johnston, 1987).

**Table 1 – Reference conditions**

<table>
<thead>
<tr>
<th>Parameters</th>
<th>Reference values</th>
<th>Investigated range of variation</th>
</tr>
</thead>
<tbody>
<tr>
<td>Particle diameter, $d_p$</td>
<td>$10^{-4} - 10 \mu m$</td>
<td>-</td>
</tr>
<tr>
<td>Dimensionless droplet charge, $q_g/q_R$</td>
<td>0 - 0.3</td>
<td>-</td>
</tr>
<tr>
<td>Droplet diameter, $D_g$</td>
<td>400 $\mu m$</td>
<td>50 - 600 $\mu m$</td>
</tr>
<tr>
<td>Droplet numerical concentration, $C_g$</td>
<td>$0.5 \cdot 10^7 m^3$</td>
<td>$1.5 \cdot 10^5 - 1.5 \cdot 10^7 m^3$</td>
</tr>
<tr>
<td>Contact time, $t$</td>
<td>3 s</td>
<td>-</td>
</tr>
<tr>
<td>Dimensionless drop-gas relative velocity, $U_g/U_t$</td>
<td>1</td>
<td>0 – 2</td>
</tr>
<tr>
<td>Volumetric volume fraction, $\varepsilon$</td>
<td>$2 \cdot 10^{-4}$</td>
<td>$10^{-7} - 6 \cdot 10^{-4}$</td>
</tr>
</tbody>
</table>

First of all, Figure 2 shows that the charging of scrubbing particles increases the collection efficiency for particles ranging between 10 nm to 10 $\mu m$. The effect is more valuable in proximity of the Greenfield gap (0.1-1 $\mu m$), confirming the potentiality of wet electrostatic scrubbing respect to traditional equipment for dust depuration.

![Figure 2 – Effect of droplet charge on the collection efficiency for different particle diameters](image)

As evident from Eq. (7) the collection efficiency exponentially increases with the contact time. This result is shown in Figure 3 that reports the time, $t_{95\%}$, required to reach 95% collection efficiency for a given charge level normalized on the time.
required without any charge for different particle sizes. The electrostatic interactions reduce by an order of magnitude the required contact time, and a charge loading level around 20% allows a contact time around the 1% of the corresponding value in a wet scrubber. A further increase of the charge levels appears to be less relevant. The efficiency improvement of a WES system is higher for finer particles due to the higher effects of electrostatic collection mechanisms (Figure 2).

![Figure 3](image-url)  
*Figure 3 - Normalized contact time for 95% collection efficiency as a function of dimensionless charge.*

The effect of the dimensionless relative velocity (given by the ratio of $U_g$ with the drop terminal velocity $U_t$), water volumetric fraction and droplet diameter are described in Figure 4 for two particle diameters representing the borders of Greenfield gap: 100 nm on the left; 1 µm on the right.

Figure 4 shows a negligible effect of relative velocity while, as expected, the collection efficiency is higher for greater water fractions. Finally, the collection efficiency increases by decreasing the droplet size. Hence, the model suggests the use of a nozzle forming well dispersed and fine droplets, apart from their velocity.

Since the capture efficiency results almost independent from relative velocity, the co-current, counter-current or crossed flows are equivalent under a theoretical point of view and the assessment of the best droplet-gas relative flow depends on different considerations. On the one hand, the counter-current flow favours a droplet dragging toward the electrical charging section and should be avoided to reduce the risk of electric discharges near the nozzle sections. On the other hand, cross-flow configurations have a similar, even if reduced drawback, but in these systems there is an enhanced risk of fluid bypass through the charging nozzle sections, giving rise to a reduction of collection efficiencies. As a consequence, co-current flow is supposed to be the best choice.
Figure 4 - Collection efficiency in function of process parameters
4. WES applications

4.1. Diesel particle removal modelling

In this section the theoretical model for particle scavenging by electrified water spray is coupled with a rapid mixing approach model (Miller et al., 1998) for droplet evaporation, in order to estimate the particle concentration after the WES treatment of a known diesel engine exhaust, as the one studied by Matti-Maricq (2006). The author reported the distribution functions of particle size and electric charge in the exhaust gases emitted by a diesel car engine at two velocity regimes. The average particle size is around 50 nm, while natural charges are between 1 and 4 electrons per particle. The only exception to Matti-Maricq’s characterization is that we “pre-conditioned” diesel exhausts to increase the relative humidity from circa 5% to 50% in order to reduce the evaporation rate of the sprayed droplets.

Following the indications reported in the former paragraphs, the WES process was applied under the following working conditions:

- Sprayed droplet size, $D = 100 \mu m$;
- Gas-droplet relative velocity, $U$, equal to the droplet terminal velocity, $U_t = 0.26 m/s$;
- Water fraction, $\Phi = 100 ml/m^3$;
- Treatment time, $t = 3 s$;
- Droplet charge, $q = 0-30\%$ of the Rayleigh limit charge $q_R$.

Figure 5 reports the number of emitted particles per Km before and after the WES treatment, at different droplet charge levels. The particle concentration after the WES treatment reduces with the exponent of the droplet charge. The model results show that theoretical removal efficiencies is around 50% for 300 nm particles, increases up to 75% for 150 nm particles and finally reaches 99.9% for finer ones.

Further increase of removal efficiency, especially for the coarser particles, can be reached by pre-charging them with corona discharge electrodes. For example, if a particle coarser than 150 nm accepts an additional charge up to 10 electrons, its concentration will decrease of two order of magnitude, as shown in Figure 6.

![Figure 5 – Emission of particulate from a diesel engine before and after WES treatment at different droplet charge levels.](image-url)
4.2. Particle removal in a WES prototype - DEECON project

This paragraph presents experimental results on the particle capture efficiency achieved in a wet electrostatic scrubber, equipped with a specially designed corona source and induction spray nozzle to generate a stream of charged particles and a spray of droplets with opposite polarities. This unit was specifically designed and constructed for marine diesel engine emission control within the activities of the 7EFP project DEECON – Innovative Technologies for Marine Diesel Engine Emission Control. Experiments were carried out on a model combustion gases with fine and ultrafine particles ranging from 10 to 1000 nm.

Wet electrostatic scrubbing tests were carried out in a pre-pilot scale system sized to treat 200 Nm³/h of gas. The WES chamber was a stainless steel column 400 mm ID and 3500 mm height equipped with a pneumatic spray nozzle electrified by induction (EFS) and with a particle charging unit (PCU) that made use of a corona source to ionize the gas and charge the particles. The gas and the spray streams flowed co-currently in the scrubber.

The source of particles was a free flame of gasoline generated with a naphtha lamp. The flue gas were aspirated by an exhaust fan and contained around 10¹² particles/m³ having size ranging between 10-1000 nm and with average diameter around 180 nm. Tests were carried out by spraying tap water with an operating liquid-to-gas mass ratio of 1.13 kg/kg. Two levels of charging potentials, 0 and 15 kV were used for the electrified spray nozzle. The same values were used for the particle charging unit.

The analysis of particulate matter was carried out with a TSI Nanoscan 3910 and a TSI LAS 3340 devices operated in parallel. The analytic system included a pretreatment section composed by thermodenuder, a 1:1000 dilutor and a particle neutralizer. The TSI 3910 allows measuring particles in the range 10-420 nm, while the TSI 3340 covers a particle size range between 90 and 7500 nm. The two devices were used to determine particle concentration and particle size distribution (PSD) in a fixed sampling point of the experimental set up, placed at the exit of the WES chamber, before the exhaust fan.
These data were eventually used for the determination of particle removal efficiency in each particle size. The instrumentations also provided the total, numerical, particle concentration, from which overall particle removal efficiency can be derived. Figure 7 describes the particle size distribution generated by the gasoline flame. The figure shows that the particulate matter is bimodal, with total concentration almost constant and close to $10^{12} \#$/m$^3$ and an average particle diameter of 190 nm.

When the WES chamber is operated with an uncharged spray and particles, the particle removal efficiency $\eta$ was very limited, being above 10% for particles finer than 220 nm, but resulted almost null for coarser particles (Figure 8). The corresponding value of the overall particle removal efficiency is around 5%.

The scrubbing efficiency increased when particle size decreased from 200 to 60 nm due to Brownian diffusion capture mechanisms. However, the values of particle abatement efficiency at 20 and 30 nm did not follow this trend.

No appreciable variation of the average particle diameter was observed in these experimental conditions.
When both the spray and the particles are charged, the particle abatement efficiency presented a dramatic increase, as shown in Figure 9. In this case, particle abatement efficiency was very high, reaching values close to 95% for particles of 300 nm and resulting higher than 80% for particles in the range between 20 and 1000 nm. The overall abatement efficiency was about 93% and the particle diameter reduced to about 160 nm.
5. Conclusions

This paper shows modelling and experimental evidences on the WES processes. A simplified model for particle capture, based on theoretical analyses of droplet-particle interactions, shows that the electrostatic charging of the droplet spray is a reliable method to enhance the collection efficiencies of wet scrubber for submicronic particles above 99%: A droplet charging around 20% results to be an optimal value, since it allows a significant reduction of treatment time and contains the electrical consumes due to the droplet charging process. The theoretical efficiencies become closer to unit by decreasing the droplet size almost independently on their velocities.

Process modelling indicates that the wet electrostatic scrubbing should reduce diesel particle concentration at levels able to comply with current EURO 4-5 regulations, while experimental tests demonstrated that the process allow reaching 93% removal in number (99% in mass) of particles finer than 1000 nm by using L/G ratio around 1.13 kg/Nm³.

Experiments and modelling highlight the potentialities of the WES processes in removing submicron particles.

Further research will bring to design a pilot scale flue gas treatment system to be installed and tested on combustion gas coming from an industrial plant. Final aim is the industrialisation of the process after optimisation for industrial applications.

References

(4) W. Licht, (1988). Air Pollution Control Engineering: Basic Calculations for Particulate Collection, 2nd ed. Marcel Dekker New York,