Wet electrostatic scrubbing for flue gas treatment

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Today SO$_2$ emissions are still too high

SO$_2$ emissions 2018: 70 MT/year
Reduced by 60 MT/year respect to 1990 – Flue gas desulphurization processes

Further reduction on SO$_2$ emissions indicated in the EU2016/2284 Directive.
China and USA introduced new restrictions recently.
The highest removal efficiencies for SO$_2$ can be achieved with Wet scrubbing systems.
The metrological problem of particle toxicity

In terms of chemical compositions, the most toxic particles are not the heavyweights!

Particles toxicity is proportional to their surface area

To reduce DPM toxicological effects, we should control the particle surface concentration. This is complex to measure even in specialised laboratories!

Simultaneous control of mass and number concentration is robust and reliable (Euro VI, 2016/1628/EU...).
Wet scrubbing for particle collection

- MULTI-CYCLONE
- VENTURI SCRubber
- WET SCRUBBER
- ESP
- FABRIC FILTER
- BUBBLE COLUMN

**ESP limitation:** depending on particle resistivity, liquid sticky aerosol

**FF limitation:** clogging, liquid sticky aerosol

Technology improvements for particles in the Greenfield gap and ultrafine range
Wet scrubbing for FGD applications
Wet scrubbing for FGD applications

Step 1- CFD code for droplets trajectories and 1D column discretization

Step 2- Estimation of $L_k$

Step 3- Mass balance equations

$G(y_k - y_{k-1}) = L_k(x_k - x_{k-1}) + L_{k,w}x_{k-1}$

Step 4- Mass transport equations

$G(y_k - y_{k-1}) = \sum_{j=1}^{N} pdf(D_j) \cdot N \cdot \frac{L_k}{L_0} \cdot K_{y,j}S_j \cdot (y_{k-1,j} - y_{k,e,j}^*)$

Step 5- Determination of outflow concentration, $x_B$, $y_B$

Droplet size distribution, pdf ($D_j$), available from experiments

$y_{k,e,j}^* = m(D_j) x_{k-1}$

Wet Scrubber & Wet Electrostatic Scrubber

**WS - Wet Scrubber**

- Efficient with COARSE particles
- Water
- Gas with PM

**WES - Wet electrostatic scrubber**

- Efficient with FINE particles
- ULTRAFINE particles?
- Water
- Gas
Wet Electrostatic scrubbing (WES) in Nature

The wet electrostatic scrubbing is based on the occurrence of electromagnetic interactions triggered by droplet charges

**OPES** – Opposite polarities electrostatic scrubbing

**CDES**- Charged droplets electrostatic scrubbing

«Nature does nothing in vain». Aristoteles
Aim of the Work

Analyse the performances of seawater-based WES units for SO\textsubscript{2} and particle removal in combustion plants
WES for sulphur dioxide capture
SO$_2$ capture in WES – Our pilot scale evidences

**DEECON twin columns set up in Napoli (IT).**

Model flue gas (3.5% S fuel)
WES: L = 195 L/h, S
WS: L = 1500 L/h.
G = 120 Nm$^3$/h
Sea water

Hydraulic nozzle with Induction charging:
$\text{q/m} = 220 \text{ mC/kg}$, Avg. droplet size = 280 $\mu$m

Corona source operating at $\approx 3 \times$ onset corona voltage

Di Natale et al. (IJHT 2016)
**SO₂ capture in WES – Our pilot scale evidences**

*DEECON testing facility in Southampton (UK)*

- Marine diesel engine 2%S IFO fuel
- Seawater
- Hydraulic nozzle with Induction charging: q/m = 150 μC/kg,
- Avg. droplet size = 250 μm
- Corona source operating at 2 x onset corona voltage

\[ \xi = \frac{y_0 - y}{y_0 - y_{eq}} \]

**Fractional approach to equilibrium**

- L/G = 2 L/m³
- OPES
  - \( \xi = 97\% \)
  - \( \xi = 91.8\% \)
  - \( \xi = 90.2\% \)
  - \( \xi = 87.8\% \)

<table>
<thead>
<tr>
<th>L/G</th>
<th>PCU</th>
<th>Voltage</th>
<th>Fractional Approach</th>
</tr>
</thead>
<tbody>
<tr>
<td>ES 0 kV</td>
<td>PCU 0 kV</td>
<td>82%</td>
<td></td>
</tr>
<tr>
<td>ES 10 kV</td>
<td>PCU 0 kV</td>
<td>86%</td>
<td></td>
</tr>
<tr>
<td>ES 0 kV</td>
<td>PCU 15 kV</td>
<td>90%</td>
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</tr>
<tr>
<td>ES 10 kV</td>
<td>PCU 15 kV</td>
<td>98%</td>
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</tbody>
</table>
SO$_2$ capture in WES - Literature survey

a. Dou et al. (2008) – SO$_2$ removal efficiency in an electrified spray dryer as a function of applied potential

b. Matteson and Giardina (1974) – SO$_2$ mass transfer coefficient of charged water droplets as a function of the droplet surface charge;

c. Wang et al. (2013) – Removal efficiency in an electrified scrubber as a function of applied potential parametric with Ca/S ratio and initial SO$_2$ concentration;

d. Carleson and Berg (1983) – Dimensionless SO$_2$ concentration in the liquid phase as a function of the electric field applied to the spray in 200 mL/min of pure SO$_2$ gas.

Effects of drops size reduction – not measured

Increase in mass transfer rate due to increased solubility

Accelerated droplets have reduced residence time
Cocurrent WES absorption experiment @ lab scale

G = 1 L/min
C₀ = 1000 ppm balance N₂
P = 1 atm; T = 20°C
L = 2-6 mL/min
L/G = 2-6 mL/L
Distilled water
CDES unit

- Very good control of spray properties
- Electrified spray simple jet mode
- Needle to ring up
- Applied voltage = 0 – 5 kV
- Contact chamber: ID = 40 mm, Z = 200 mm
- Stainless-steel needle (OD 0.8 mm and ID 0.5 mm)
- Droplets size 1 – 2 mm Typical of FGDs

Di Natale et al, ChERD, 2019
Parameters assessment - Optical and electrical tests

- **Optical tests**
  - CMOS Photron Ultima APX camera equipped with a Nikon af-s nikkor 85 mm f/1.8 g lens
  - Frame rate of 500 fps
  - Shutter speed or exposure time of 1/10,000 s.
  - Sampling time 1 min

- **Electrical tests**
  - Spray current, I
  - Charge to mass ratio (D-CMR= q/m= I/L)

**Image analysis** allows determination of:
- Droplets’ size distributions: \(d_3\), \(d_{32}\);
- Droplets’ deformation/aspect ratio/circularity distribution
- Frequency of formation, \(f\)
- Frequency of oscillation, \(w\)

**Electrical analysis** (Keithley, mod. 6514) allows determination of:
- Spray current, I
- Charge to mass ratio (D-CMR= q/m= I/L)
Continuous Lab scale tests

Overall mass transfer coefficient

$$k_{goa} \propto \left[ \frac{L}{G} \cdot \frac{da}{dt} \right]^{0.492} \cdot q_D^{0.418}$$

Di Natale et al. (ChERD 2019)
Feed-batch system with E.S. in dripping mode with and w/o corona charging

V = 2.08 L
C₀ = 1000 ppm balance N₂
P = 1 atm; T = 20°C
L = 1 mL/min
Distilled water
OPES/CDES unit
Corona ch. -10/+15 kV
Corona onset ≈ ± 6.3 kV

- Electrified spray in dripping mode
- Needle to ring up
- Applied voltage = 0 – 2.4 kV
- Contact chamber: ID = 100 mm, Z = 200 mm
- Stainless-steel needle (OD 0.8 mm and ID 0.5 mm)
- Droplets size 1.8 – 2.2 mm Typical of FGDs

Di Natale et al, CET 2016; D’Addio et al., CES 2013, 2014
Experiments

No effects of droplets charge polarity

Negligible effects of gas charging

CDES

OPES
Absorption model for charged droplets

Gas diffusivity + Electrophoresis (?)

Liquid diffusivity

\[ J_i = -\frac{D_i c_i}{k_B T} \left( \frac{\partial \mu_i}{\partial x} + Z_i e \frac{\partial \Phi}{\partial x} \right) \]

Alteration of drop morphology and surface area
Switch to Oscillating drops

Bulk and/or Interfacial solubility
It is possible to express the Henry solubility constant as (Uhlig, 1937):

\[
\ln(m) = \frac{4\pi r^2 \gamma_0}{kT} - \frac{E}{kT}
\]

- \(r\) is the molecular radius (= 0.12391 for \(\text{SO}_2\));
- \(k\) is the Boltzmann’s constant;
- \(T\) is the absolute temperature;
- \(E\) is the interaction energy;
- \(\gamma\) the liquid surface tension.

Assuming \(E\) as constant (solvent-solute interactions), it is also possible to write the Henry Constant in presence of a surface charge \(q\) (\(V\)):

\[
\ln m(V) = \frac{4\pi r^2 \gamma(V)}{kT} - \frac{E}{kT}
\]

Being:

\[
\frac{\gamma(V)}{\gamma_0} = 1 - \left(\frac{q(V)}{q_R}\right)^2
\]

Therefore:

\[
\ln m_e = \ln m_0 \cdot \left(1 - \frac{q(V)^2}{q_R^2}\right)
\]

For \(\frac{q}{q_R} \approx 0.3\), \(\frac{m_e}{m} = 0.67 \rightarrow \text{Solubility increases by 30%!!}\)
Several models used to describe mass transfer during droplet falls to account for deformation and oscillations ($Re>60; \quad d_p>1.7 \text{ mm}$)

**Liquid-film resistance models**

<table>
<thead>
<tr>
<th>Stagnant flow</th>
<th>Circulating flow</th>
<th>Oscillating flow</th>
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<tbody>
<tr>
<td>$D_L &lt; 500 \mu m$</td>
<td>$500 \mu m &lt; D_L &lt; 1700 \mu m$</td>
<td>$D_L &gt; 1700 \mu m$</td>
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**Gas-film resistance models**

<table>
<thead>
<tr>
<th>Circulating flow models</th>
<th>Gas-film resistance models</th>
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</thead>
<tbody>
<tr>
<td>Dimiccoli et al. (2000)</td>
<td>Lochiel &amp; Calderbank (1964)</td>
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<td></td>
<td>Garner &amp; Tayeban (1960)</td>
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<td></td>
<td>Griffith (1960)</td>
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<td></td>
<td>Hughmark (1967)</td>
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<td>Saboni &amp; Alexandrova (2001)</td>
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</table>

**Oscillating droplets**

<table>
<thead>
<tr>
<th>Handlos &amp; Baron (1957)</th>
</tr>
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<tbody>
<tr>
<td>Rose &amp; Kintner (1966)</td>
</tr>
<tr>
<td>Clift et al. (1978)</td>
</tr>
<tr>
<td>Brunson &amp; Wellek (1970)</td>
</tr>
<tr>
<td>Yamaguchi et al. (1975)</td>
</tr>
</tbody>
</table>

Models predictions

Models range represents **minimum** and **maximum** predictions achievable with all the aforementioned models.

- **0 kV Sph/Uncharged**
- **1.2 kV Osc/Charged**
- **1.8 kV Osc/Charged**
- **2.4 kV Osc/Charged**
Models predictions – influence of solubility and mass transfer models
## Models predictions

### Coefficient of determination, $R^2$.

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<tr>
<th>V, kV</th>
<th>2.4</th>
<th>2.1</th>
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<th>1.2</th>
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<td>Handlos &amp; Baron</td>
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<td>81.57%</td>
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<td>80.99%</td>
<td>80.50%</td>
<td>82.66%</td>
<td>84.72%</td>
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<td>Hughmark</td>
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<td>75.16%</td>
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<td>Saboni</td>
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<td>75.00%</td>
<td>81.70%</td>
<td>74.27%</td>
<td>73.48%</td>
</tr>
</tbody>
</table>

**Clift model** for mass transfer coefficients provides the **best prediction of data**.

Residual errors are probably related to underestimation of ion diffusivity in charged droplets.
WES for particles capture
Wet electrostatic scrubber – uncharged particles

Each point corresponds to the average removal efficiency of a given particle size distribution. Different L/G ratio and WES design are explored.

Zoom on higher-flow rates: large spray nozzle (3-25 L/min)
Limited or negligible alteration of droplet size distribution.

Enhanced removal efficiency by charge density!

Zoom on lower-flow rates: small spray needle (2-10 mL/min)
Relevant alteration of droplets morphology (Dripping to Whipping modes explored)
Wet electrostatic scrubber – charged particles

Test bench facilities:
Model 1: 150 Nm3/h (2012-2013)
Model 2: 1000 Nm3/h (2014)
Model 3: <10000 Nm3/h (since 2017)
Model 4: <15000 Nm3/h (since 2016)
Model 1 - pilot-scale WES an example

- Gas distributor and scrubber head
- Quencher
- Model gas generation system
- PLC and main electric cabinet
- Gas analysis rack
- Particle charging units
- WES column
- SWS column
- WWT tank
- Particle analysis rack
- Anticontamination system electric cabinet

Emitted particles

Source of particles
Gasoline flame generated with a naphtha lamp burned in open air

Particle Size Distribution measured simultaneously by:

**TSI 3910**
Nanoscan SMPS Nanoparticle sizer 3910
Range 10-420 nm

**TSI 3340**
Laser Aerosol Spectrometer 3340
Range 90-7500 nm

![Graphs showing particle size distribution](image)

*Fig. 3. Normalized particle concentration as a function of particle diameter measured with (a) TSI 3910 and (b) TSI 3340. C = 170kg/h, T= 25°C, RH = 100%.*

Pilot-scale experimental results

UNCHARGED DROPLETS
UNCHARGED PARTICLES

CHARGED DROPLETS
UNCHARGED PARTICLES

CHARGED DROPLETS
CHARGED PARTICLES

negligible removal efficiency

Image-charge forces among particles and droplets

Coulomb attraction forces among particles and droplets

Liquid flow rate, $L = 195$ kg/h
Gas flow rate, $G = 170$ Kg/h
L/G = 1.15
Liquid Potential $V_L = 0$ kV
Gas Potential $V_G = 0$ kV

L = 195 kg/h
G = 170 Kg/h
L/G = 1.15
$V_L = -15$ V
$V_G = 0$ kV

L = 195 kg/h
G = 170 Kg/h
L/G = 1.15
$V_L = -15$ kV
$V_G = -15$ kV

Prototype bench scale tests (up to 15,000 Nm³/h) show removal efficiencies similar to that of FF or ESP. SO₂ absorption can be carried out simultaneously.
Mathematical model

...for a generic droplet size distribution, $\psi (D_g)$...

Population balance

Evolution of particle concentration

Capture efficiency
Mathematical model – effect of droplet charge
Mathematical model

**EFFECT WATER CONCENTRATION**

Fig. 7. Collection efficiency as a function of volumetric water fraction for three particle diameters, (a) 100 nm, (b) 1 μm, (c) 5 μm, and different droplet charge levels: (○) \( q_{\text{aq}} = 0 \), (●) \( q_{\text{aq}} = 0.01 \), (■) \( q_{\text{aq}} = 0.05 \), (▲) \( q_{\text{aq}} = 0.1 \), (▲) \( q_{\text{aq}} = 0.3 \). Reference conditions are reported in Table 3.

**EFFECT WATER/GAS RELATIVE VELOCITY**

Fig. 9. Collection efficiency as a function of water/gas relative velocity for three particle diameters, (a) 100 nm, (b) 1 μm, (c) 5 μm, and different droplet charge levels: (○) \( q_{\text{aq}} = 0 \), (●) \( q_{\text{aq}} = 0.01 \), (■) \( q_{\text{aq}} = 0.05 \), (▲) \( q_{\text{aq}} = 0.1 \), (▲) \( q_{\text{aq}} = 0.3 \). Reference conditions are reported in Table 3.
Comparison with experimental data

**EFFECT OF PARTICLE CHARGE**

By **increasing** the particle charge:

- The scavenging coefficient **increases**
- The model/experimental comparison **worsens**

...but the scavenging model used for micron size particles works satisfactorily also for submicron particles
Conclusions: What we understood so far

«Nature provides exceptions to every rule». M. Fuller

- Electrostatic scrubbing influences absorption of SO$_2$ with marked effects for $q/q_R < 50\%$ (+40\% @ $q/q_R < 10\%$)

- Electrifications affects both droplets morphology and interfacial conditions – both of them influences the absorption rate.

- Experiments in dripping mode can be predicted by current models

- SO$_2$ absorption is favoured by small droplet size and high charge density (not as PN!)

Further developments: Short term objectives

- Include estimation of dissolved SO2 ions diffusivity in charged droplets (Work in progress)
- Extension to other water soluble gases and water solution compositions
- Extend modelling to full scale scrubbers
Thank you for your attention!