Development of wet phase transition agglomerator for multi-pollutant synergistic removal

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Abstract: This study presents an originally designed device named as wet phase transition agglomerator (WPTA) based on vapour condensation agglomeration and collection mechanism. This technology has achieved demonstration and commercialization in several industrial / utility boilers. Excellent performances have been validated in fine particles and trace elements synergistic removal, latent heat and water recovery, and sulfuric acid mist control. The vapour condensation produced by the WPTA greatly improve the agglomeration among fine particles and subsequent removal. The removal efficiency of WPTA combined with wet electrostatic precipitator (WESP) for PM$_{1.0}$ at 600 MW was enhanced by 15%, compared to the operation at WPTA shutdown. About 13.4 ~ 15.6 t/h of condensing water was recovered when the designed flue gas temperature was decreased by 1 ~ 1.5 °C. Moreover, the WPTA was capable of significantly improving the removing amounts of soluble salts and trace elements from flue gases.

Keywords: Wet phase transition agglomerator; Synergistic removal; Fine particles; Trace elements; Vapour condensation

1 Introduction

In 2014, the ambient PM$_{2.5}$ level in China was significantly higher than the recommended value of 10 μg/m$^3$ issued by the World Health Organization [1]. It has been found that the rise of 10 μg/m$^3$ in ambient PM$_{2.5}$ leads to a 0.02 ~ 0.05 mg increase of
particles deposited in the lung per day [2]. To confront such severe atmospheric environmental problems, China’s Department of Environmental Protection has issued the most stringent air pollutant emission standard (NO\textsubscript{x} < 50 mg/Nm\textsuperscript{3}, SO\textsubscript{2} < 35 mg/Nm\textsuperscript{3}, PM < 10 mg/Nm\textsuperscript{3} even 5 mg/Nm\textsuperscript{3}), for coal-fired power plants with the generation capacity of more than 300 MW [3]. It is higher than the standards for coal-fired units in Germany (NO\textsubscript{x} < 150 mg/Nm\textsuperscript{3}, SO\textsubscript{2} < 150 mg/Nm\textsuperscript{3}, PM < 10 mg/Nm\textsuperscript{3}) [4] and is similar to that in America (NO\textsubscript{x} < 42 mg/Nm\textsuperscript{3}, SO\textsubscript{2} < 50 mg/Nm\textsuperscript{3}, PM < 6 mg/Nm\textsuperscript{3}) [5]. Accordingly, the advanced technologies for NO\textsubscript{x}, SO\textsubscript{2} and PM removal are common in Chinese coal-fired units. But they have a difficulty in achieving the synergistic removal of various pollutants, or a lower synergistic removal capacity. Therefore, studying the efficient synergistic removal technologies for multiphase contaminants is of great importance.

At present, the PM\textsubscript{2.5} control research is still focused on how to improve the agglomeration characteristic. The traditional technologies, such as electrostatic precipitators (ESP) and cyclone separators, have difficulties in efficiently removing these particles at the diameter of 0.1 ~ 1 μm. Therefore, advanced technologies are required to preprocess fine particles, and enlarge their sizes. Such technologies include chemical agglomeration [6], acoustic wave agglomeration [7], magnetic agglomeration [8], vapour condensation agglomeration [9] and turbulent agglomeration [10], etc. The core of these technologies can be classified into two aspects, i.e. (1) enhancing the collision efficiency between fine particles via extending the external physical fields; (2) altering the adhesive properties of the surfaces of impacting particles. At present, the WESP acting as a finer processing device has been employed in many Chinese coal-fired power plants. This technology could accomplish the synergistic removal of fine particles, Hg and SO\textsubscript{3}, even though its capital investment and removal cost are extremely expensive. As a result, it is necessary to seek the strategies for operating costs reduction and to evolve alternative pretreated technologies for environment-friendly technology development and improvement.

The previous studies regarding the vapour agglomeration technology mainly used the method of spraying water vapours to enhance the condensing growth on the surface of fine
particles [11]. But in this process, fine particles could not be efficiently transported toward the surface of condensing water. Most of them could move in the opposite direction, due to the higher temperature of spraying water vapours than that of fine particles. Actually, the practical way to enhance the removal efficiency of fine particles is to use a condensing heat exchanger to condense water vapor out from wet flue gas. In this process, lots of contaminants could be removed by the cooled pipes and condensing water, but the acid dew corrosion and fouling resistance of metal pipes have to be considered [12,13]. In this study, an agglomeration technology was designed and proposed on a basis of vapour condensation agglomeration and collection mechanism. The key design concepts, and technical development as well as removal mechanism of multi-pollutant for the WPTA technology were elaborated in detail. Moreover, the removal capacity of the WPTA for fine particles, trace elements and inorganic ions, and the performance of latent heat and water recovery were analyzed.

2 Technical development of WPTA

It is well-known that, during the raindrop settling process, they are able to capture some of the fine particles suspending in the air. Based on this principle, the WPTA device was designed and developed. It utilizes the feature of cross flow heat exchanger, to make the wet flue gas cooled down. In the WPTA device, the condensate always covers the tube surfaces and alters its properties. The transporting process of condensed water mist towards the tubes, could also capture some fine particles. In addition, the water vapour reacts with SO$_3$ to produce the sulfuric acid mist, which dissolves some soluble inorganic salts on the surface of fine particles. Some trace elements could also be removed by the WPTA. Several experiments [14,15] found that trace elements such as Hg, As and Cr are enriched in submicron particles. Flowing across the tube bundles inside the WPTA, fine particles could be effectively captured by water mist, or removed by the condensed film.

The WPTA device is fully made up of perfluoroalkoxy (PFA) material [16], which can overcome the corrosion and fouling. Furthermore, it has higher upper-temperature limits [17], and can be expanded into more extensive fields. Fig. 1 presents the pictures of WPTA setup.
from lab-scale to full-scale. The lab-scale tests of the WPTA have been completed in 2011, as present in Fig.1(a), aiming to determine the optimal dimensions and the performances of water and heat recovery. From 2012 to 2013, the pilot-scale tests of the WPTA were conducted in a 600MW lignite-fired power plant, as seen in Fig.1(b). The full-scale demonstration of WPTA in a 660 MW coal-fired power plant was completed at the end of 2014 and was installed at the inlet of the WESP equipment (see Fig.1(c)). It combines with WESP configuration to reduce the total mass concentration of ash particles at the outlet to be lower 5 mg/m$^3$. Fig.1(d) shows a modularized WPTA unit with a height of 600 mm. Its dimension is flexible according to different installation position. More information about the WPTA equipment can be seen in elsewhere [16, 18, 19]. Table 1 gives a comparison of some of the key parameters related to the WPTA. This device has a low operation resistance and a small influence on the up-down stream equipment. Currently, several industrial/utility boilers have installed the WPTA device to achieve the ultra-low emission of fine particles.

![Fig. 1. Development and application of WPTA (a-lab-scale; b-pilot-scale[16,18]; c-full scale boiler; d-WPTA module)](image)

**Table 1**
Some of the key parameters for WPTA.

<table>
<thead>
<tr>
<th>Item</th>
<th>Unit</th>
<th>Lab-scale test</th>
<th>Pilot-scale test</th>
<th>Full-scale test</th>
</tr>
</thead>
<tbody>
<tr>
<td>Flue gas flow</td>
<td>m³/h</td>
<td>1,728</td>
<td>29,584 ~ 60,212</td>
<td>2,780,666</td>
</tr>
<tr>
<td>Flow cross-section</td>
<td>m²</td>
<td>0.6 × 0.12</td>
<td>1.5 × 1.38</td>
<td>14.43 × 0.95</td>
</tr>
<tr>
<td>Stage</td>
<td>/</td>
<td>One</td>
<td>Two</td>
<td>One</td>
</tr>
<tr>
<td>System resistance</td>
<td>Pa</td>
<td>200 ~ 900</td>
<td>121 ~ 430</td>
<td>30</td>
</tr>
<tr>
<td>Inlet flue gas velocity</td>
<td>m/s</td>
<td>3 ~ 8</td>
<td>3.6 ~ 8.6</td>
<td>&lt; 3</td>
</tr>
<tr>
<td>Inlet flue gas temperature</td>
<td>°C</td>
<td>45 ~ 55</td>
<td>55 ~ 57</td>
<td>50 ~ 57</td>
</tr>
</tbody>
</table>

3 Theory analysis of fine particle removal

When the flue gas temperature decreases, vapour condenses into a large number of small water drops, greatly enhances the collision probabilities among fine particles. This condensation effect also promotes some of the fine particles moving towards the droplets. In order to understand the removal characteristics of water mist for fine particles, and to evaluate the impaction efficiency between water mist and ash particle at different scales a dimensionless semi-empirical relationship is introduced [20],

\[
E(d, D) = \left\{ \frac{4}{ReSc} \left[ 1 + 0.4Re^{1/2}Sc^{1/3} + 0.16Re^{1/2}Sc^{1/2} \right] \right\}_{\text{Brownian diffusion}} + \left\{ \frac{4d}{D} \left[ \frac{\mu_w}{\mu} + \left( 1 + 2Re^{1/2} \right) \frac{d}{D} \right] \right\}_{\text{interception}} + \left\{ \left( \frac{\rho_w}{\rho_p} \right)^{1/2} \left( \frac{St - S^*}{St - S^* + 2/3} \right)^{3/2} \right\}_{\text{inertial impaction}} ,
\]

where the dimensionless parameter \( S^* \) is calculated by the expression \((1.2 + (1/12)\ln(1+Re))/(1+\ln(1+Re))\). On the right side of Eq.(1), the impaction efficiency is mainly caused by the Brownian diffusion, interception and inertial impaction. Here two diameters (0.5 and 1.5 mm) of water droplet were assumed to calculate and to emphasize which effect is dominant at different size. As presented in Fig.2, the contribution of Brownian diffusion to the impaction efficiency is gradually decreased with increasing the particle diameter. The inertial impaction has a dominant influence on the coarse particles, but the effect of Brownian diffusion and inertial impaction on impaction efficiency between droplets and particles at the diameter of 0.01 ~ 1 μm was pretty small. Additionally, the interception effect of droplet on particles at the size of 0.01 ~ 1 μm can be neglected.
Fig. 2. Impaction efficiency between water droplet and ash particles

However, the effect of temperature gradient on impaction efficiency was not taken into consideration in the semi-empirical relationship. During the cooling process of flue gases, it has a dominant role on the motion of fine particles [21]. The thermophoretic force upon a particle is induced by the local temperature gradient [22], as follows,

$$
F_{th} = \frac{1.15 Kn \left(1 - \exp \left(-\frac{a}{Kn}\right)\right) \frac{4}{3\pi} \phi \pi Kn k_B d^2}{4\sqrt{2a} \left(1 + \frac{\pi_1}{2} Kn\right)} \nabla T,
$$

(2)

where the Knudsen number $Kn$ is defined as the ratio of the mean free path to the particle radius. In Eq (2) the calculations for these variables $\phi$, $a$, $\pi_1$ are given by

$$
\phi = 0.25 (9\gamma - 5) C_v / R_g
$$

(3)

$$
a = 0.22 \left(\frac{\phi \pi / 6}{1 + Kn \pi_1 / 2}\right)
$$

(4)

$$
\pi_1 = 0.18 \frac{36/\pi}{(2 - S_n + S_\pi) 4/\pi + S_n}
$$

(5)

The thermophoretic force was calculated from Eq.(2) based on the absolute pressure of 0.1 atm and flue gas temperature of 330 K. The dimensionless thermophoretic force is presented as a function of particle size in Fig.3. The thermophoresis has a remarkable effect on the transport of fine particles (diameter of 0.01 ~ 1 μm) to the surface of pipes, in comparison
with that acting on coarse particles. Hence the effect of thermophoretic force on fine particle motion should be considered.

![Thermophoretic force graph](image)

**Fig. 3.** Thermophoretic force

### 4 Result and discussion

#### 4.1 Pilot-scale test

The ash particles were collected at three positions – the inlet and outlet of the first stage WPTA, and the outlet of the second stage WPTA. Firstly, the samples were dispersed in deionized water, and then the associated ultrasonic was enabled to improve the dispersibility. After the pretreatment, the sizes of samples were analyzed by the Masterizer 2000 laser particle size analyzer with the wet samples injection. Fig.4 shows the analytic results of three ash samples. The lines with symbols in Fig. 4 represent the particle volume at different diameter, while the double dotted lines denote the accumulated fraction of particle volume.

The particle volume distribution at the inlet of the first stage WPTA presents a unimodal peak. After flowing through the first stage WPTA, the peak value decreased sharply, and a new peak appeared at the diameter over 10 μm. This implies that the condensation of water vapour promotes fine particles to be agglomerated together. But when the flue gases flowed through the second stage WPTA, the peak value of the particle volume fraction at about 20 μm was decreased, and the former peak corresponding to the diameter of 2 μm was
significantly increased. During the cooling process of flue gases, the agglomeration and removal behaviour of the ash particles exist in parallel. In the first stage, high number concentration of fine particles results in high impaction frequency, so the agglomeration effect of fine particles takes a dominant role in particle removal. However, in the second stage, the increasing number concentration of coarse particles further improves its removal. The characteristic diameters $D_{0.1}$, $D_{0.5}$ and $D_{0.9}$ respectively correspond to the accumulated volume fractions 10 %, 50 % and 90 %, as given in Table 2. At the outlet of the first stage WPTA, the characteristic diameters $D_{0.5}$ and $D_{0.9}$ increased significantly to 18.8 μm and 101.5 μm, respectively. This also suggests the WPTA enhances the agglomeration of fine particles.

![Fig. 4. Particle volume distribution at different size](image)

**Table 2**

Particle diameter at different accumulated volume fraction.

<table>
<thead>
<tr>
<th>Item</th>
<th>Inlet</th>
<th>Outlet</th>
<th>Outlet</th>
</tr>
</thead>
<tbody>
<tr>
<td>$D_{0.1}$</td>
<td>1.12</td>
<td>1.62</td>
<td>1.38</td>
</tr>
<tr>
<td>$D_{0.5}$</td>
<td>3.54</td>
<td>18.82</td>
<td>6.09</td>
</tr>
<tr>
<td>$D_{0.9}$</td>
<td>21.36</td>
<td>101.48</td>
<td>47.06</td>
</tr>
</tbody>
</table>

**4.2 Removal efficiency of WPTA combined with WESP for fine particles**

The Dekati Low Pressure Impactor was used for sampling the ash particle at the outlet of WESP. The effects of unit load and the WPTA operation state (on and off) on particle size distribution (PSD) were investigated. The continuous sampling time was 30 min for each case. Before collecting ash particles, the sampling probe was pre-heated to about 110 ~ 120 °C in
order to reduce the condensation of water vapour in the flue gas. The round aluminium foil with turpentine coating was fixed at each impactor to capture the ash particles. The mass of the collected particles at each stage was obtained by weighing the aluminium foil.

The effect of WPTA operating state on the PSD was tested at the unit load of 500 and 600 MW, as shown in Fig. 5. Under the same unit load of 600 MW, when the WPTA was enabled, the concentrations of particle mass at the sizes of 0.03, 0.05, 0.06, 0.1, 0.47, 8.28 and 22 μm respectively exhibited different decreasing amplitudes, in contrast with those obtained at the state of WPTA off. The mass concentration of ash particle corresponding to 0.05 and 0.1 μm was respectively decreased by 57.6 % and 70.2 %. Similarly, under the unit load of 500 MW, the substantial reduction in particle mass concentration at several particle sizes such as 0.03, 1.9 and 3.1 μm was also observed in Fig.5. The mass concentration of particle at these sizes was decreased by 80 %, 35.5 % and 53.6 %, respectively. The effect of WPTA operation state on removal efficiency for PM_{1.0}, PM_{2.5} and TSP (total suspended particulates) was given in Fig. 6. When the WPTA device was enabled at 600MW, the removal efficiency for PM_{1.0}, PM_{2.5} and TSP has been increased by approximately 15 %, 5 % and 4 %, respectively. Similarly, in case of 500 MW unit load, the running WPTA could also enhance the removal efficiency for PM_{1.0}, PM_{2.5} and TSP by about 2.5 %, 3.3 % and 3.2 %, respectively. These results could prove that the WPTA has an excellent ability in fine particle removal.
4.3 Recovery capacity of condensing water from flue gas

There is a great potential to recover water vapour and latent heat from wet flue gases [23,24]. For example, in a 600 MW lignite-fired power plant, the moisture flow rate accounts for about 16 % of the flue gases [25]. During the cooling process of the WPTA, the water vapour flows through the staggered tube bank, either homogeneously condensed into water mist or heterogeneously condensed on the pipe or particle surface. The recovered condensation water volume could be obtained via recording the level line in the tank. When the WPTA is off, the recovered condensation water flux was approximately 2 t/h at 400 MW. When the WPTA was enabled, it was improved significantly, as shown in Table 3. The operational WPTA could decrease the flue gas temperature by about 1 ~ 1.5 °C. These results suggest that the WPTA device has a good performance in condensing water recovery. In addition, the responding latent heat could also be recovered. For the pilot-scale test, the maximum recovered heat of 92 MWth accounts for about 14 % of the generated thermal capacity of 660 MW [16]. In the large-scale test the recovered heat was directly released into the air, due to the problems of space, heat exchange network complexity and investment.

Table 3
Recovered mass of condensed water when the WPTA is enabled.
<table>
<thead>
<tr>
<th>No.</th>
<th>Unit load</th>
<th>Average inlet flue gas temperature (°C)</th>
<th>Water recovery mass (t/h)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>390</td>
<td>52.5</td>
<td>13.36</td>
</tr>
<tr>
<td>2</td>
<td>400</td>
<td>51.8</td>
<td>14.00</td>
</tr>
<tr>
<td>3</td>
<td>400</td>
<td>51.5</td>
<td>15.59</td>
</tr>
<tr>
<td>4</td>
<td>400</td>
<td>51.6</td>
<td>13.84</td>
</tr>
<tr>
<td>5</td>
<td>603</td>
<td>54.2</td>
<td>13.36</td>
</tr>
<tr>
<td>6</td>
<td>603</td>
<td>53.5</td>
<td>14.79</td>
</tr>
<tr>
<td>7</td>
<td>607</td>
<td>53.9</td>
<td>14.79</td>
</tr>
<tr>
<td>8</td>
<td>605</td>
<td>54.9</td>
<td>15.59</td>
</tr>
<tr>
<td>9</td>
<td>600</td>
<td>53.6</td>
<td>14.95</td>
</tr>
<tr>
<td>10</td>
<td>600</td>
<td>54.6</td>
<td>14.79</td>
</tr>
<tr>
<td>11</td>
<td>600</td>
<td>54.9</td>
<td>14.95</td>
</tr>
</tbody>
</table>

4.4 Removal capacity of WPTA for trace elements

When the WPTA was enabled or not, the condensate was collected before entering the wastewater tank. The Hg and As contents in the sampled water were detected by XGY-1011A atomic fluorescence spectrophotometer. Other elements Ba, Ga, Li Mn, Sr and Ti in the samples were detected by an inductively coupled plasma-optical emission spectroscopy (Perkin Elmer, USA). Based on the condensate volume in the WPTA operation or shutdown, the total removal amount of each trace element is defined as the volume of recovered water per hour, multiplying by the concentration of each trace element in the sample. Fig. 7 presents the total removal amount of each trace element. Obviously, the enabled WPTA could greatly improve the trace element removal.
Fig. 7. Effect of WPTA state on total removal amount of each element

Here a parameter \( W \) is proposed to evaluate the removal capacity of WPTA for each trace element, as defined by,

\[
W = \frac{m_t - m_0}{m_0},
\]  

Fig. 8 presents the removal capacity of WPTA operation for tested elements. The removal capacity of WPTA operation for Ba, Hg and As was respectively enhanced by 7.17, 4.18 and 2.82 times, in contrast with those obtained with the WPTA off. The corresponding removal capacity is listed in order: Ba > Hg > Sr > Ga > As > Ti > Li > Mn.

![Removal capacity of the WPTA for testing elements](image)

**Fig. 8.** Removal capacity of the WPTA for testing elements

4.5 Quality analysis of recovered condensation water

Table 4 presents the quality of the recovered water. At a down state of WPTA, the ion concentrations in sampled water were larger than those obtained at its normal operation. This is attributed to the significant increase in the condensing water volume. Once the WPTA device was enabled, the condensate flux was seven times greater than that with the WPTA disabled. Accordingly, the conductivity and salinity of the condensate were decreased by two times, in comparison with those at the shutdown of WPTA. Calculations with assuming the same condensate density, showed the total removal amounts of each ion were substantially increased when the WPTA was enabled.
However, the recovered water has a high PH value no matter whether the WPTA device is enabled or not. This suggests large amounts of SO$_2$ or SO$_3$ dissolved into the water. The removal amount of SO$_4^{2-}$ in the recovered condensation water was improved by approximately 4.4 times when the WPTA was enabled. The result further proves the excellent performance of WPTA in term of SO$_2$/SO$_3$ and sulphates removal. The recovered water can be used as the supplemental makeup water for wet FGD system. The limestone wet FGD method has a largest water consumption [26], so water recovery from flue gases is of great significance. However, the necessary treatments such as neutralization reaction and chemical precipitation, should be conducted before utilizing them.

5 Conclusions

A novel WPTA device was originally designed, developed, demonstrated and commercialized for industrial and utility boilers. Based on the experimental results, excellent performances of this technology were summarized in these aspects:(1) effective agglomeration capacity for PM; (2) combining with WESP, improve the removal efficiency of PM$_{2.5}$ and PM$_{1.0}$ by 5 % and 15 % at 600 MW, respectively; (3) improve significantly the removal amounts of soluble salts (Ca$^{2+}$, SO$_4^{2-}$, Cl$^-$, etc.) and trace elements (Hg, As, Ba, Ga, etc.); (4) achieve latent heat and water recovery from flue gases. It has a remarkable potential in reducing air pollutant emissions and relieving haze weather in future. More potential benefits will also be obtained if the cooling effect of flue gases is increased.
Acknowledgments

We greatly thank to the support by the National Natural Science Foundation (No. 91544108).

Nomenclature

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>$d$</td>
<td>diameter of particulate matter, m</td>
</tr>
<tr>
<td>$d_m$</td>
<td>average gas molecular diameter of mixing N$_2$ and CO$_2$ gases, $\approx 0.161$ nm</td>
</tr>
<tr>
<td>$D$</td>
<td>diameter of water droplet, m</td>
</tr>
<tr>
<td>$k_B$</td>
<td>Boltzmann constant, $1.38 \times 10^{-23}$ a flue gas</td>
</tr>
<tr>
<td>$Kn$</td>
<td>Knudsen number</td>
</tr>
<tr>
<td>$m$</td>
<td>the amount of each tested element in total recovered water, kg/m$^3$</td>
</tr>
<tr>
<td>$Re$</td>
<td>Reynolds number based on the water droplet diameter</td>
</tr>
<tr>
<td>$R_g$</td>
<td>gas constant of air, 287 J/(kg·K)</td>
</tr>
<tr>
<td>$Sc$</td>
<td>Schmidt number</td>
</tr>
<tr>
<td>$St$</td>
<td>Stokes number with the particle diameter as characteristic length</td>
</tr>
<tr>
<td>$S$</td>
<td>adjustable coefficient, $S_n=S_t=1.0$</td>
</tr>
</tbody>
</table>

Subscripts

- 0 at closed state of WPTA
- 1 at operation state of WPTA
- a flue gas
- p particulate matter
- n perpendicular direction
- t tangential direction

Abbreviations

- ESP electrostatic precipitation
- FGD flue gas desulfurization
- PFA perfluoroalkoxy
- PM particulate matter
- PSD particle size distribution
- TSP total suspended particulates
- WESP wet electrostatic precipitation
- WPTA wet phase transition agglomerator

Reference


