CHEMICAL ENGINEERING TRANSACTIONS

VOL. 52, 2016

A publication of AIDIC
The Italian Association
of Chemical Engineering
Online at www.aidic.it/cet

Guest Editors: Petar Sabev Varbanov, Peng-Yen Liew, Jun-Yow Yong, Jiří Jaromír Klemeš, Hon Loong Lam

ISBN 978-88-95608-42-6; **ISSN** 2283-9216

A Heterogeneous Condensation Assisted Three-Phase Bed Column to Remove Submicronic Particles

Francesco La Motta*[a], Francesco Di Natale*[a], Claudia Carotenuto[b], Amedeo Lancia[a]

[a]University of Naples, Department of Chemical, Material and Production Engineering, P.le Tecchio 80, 80125 Napoli,Italy
[b] Dipartimento di Ingegneria Industriale e dell'Informazione, Seconda Università degli Studi di Napoli,Aversa, Caserta; Italy
francesco.lamotta@unina.it

The paper presents experimetnal findings on an innovative system aimed to remove ultrafine particles from gases. The system is based on coupling a particle enlargement process with a three-phase bubble column. Calibrated polystyrene nanoparticles of about 200 nm were tested and were enlarged in a growth-tube operated at different temepartures to obtain different super-saturation levels. The experimental results showed that the aerosol cumulative distribution functions shifted towards larger diameters with the increasing of supersaturation; the total particle removal efficiency in the three-phase bubble column increased with the supersaturation reaching values of 99 %.

1. Introduction

The emission of particulate matter from industrial and vehicles exhausts is a major health and environmental concern (Menon et al., 2002). In particular, the submicronic (dp<1 μm) and ultrafine (dp<100 nm) particles remain suspended in the atmosphere for a long time being transported at long-distances. The reduction of visibility in the cities and scenic areas, the large climate forcing effects of Black Carbon or the solar radiation absorption are some consequences of the particulate matter on the environment (Kerr, 2013). Even more importantly, the ultrafine particles, once inhaled, can reach the deepest regions of the lungs and even enter in the circulatory system, causing a wide range of health problems (Di Natale and Carotenuto, 2015), as lung cancer (Pope et al., 2002).

The effective removal of ultrafine particles is receiving growing attentions, pushing forward the development of new processes (D'Addio et al., 2013) and technologies (Di Natale et al., 2015). In fact, the traditional particle abatement devices are mainly designed and optimised to treat particles with sizes above 2 μm, and they are far less efficient in collecting sub-micrometric particles, especially those in the range 0.1–2 μm, also known as Greenfield gap (Seinfeld and Pandis, 1998). In this paper, we are presenting a new kind of particle removal device based on the concept of bubble columns. A bubble column reactor is basically a cylindrical vessel with a gas distributor at the bottom. The gas bubbles into either a liquid phase or a liquid-solid suspension.They are widely used in chemical process industries (Kantarci et al., 2005), as bubble reactor (Yao et al., 2014) for its various advantages and simplicity. Recently several authors studied them as alternative removal technique for capture of fly-ash (Meikap and Biswas, 2004) and nanoparticles (Charvet et al., 2011).

It was highlighted that the removal efficiency of micronic particles increased with the liquid level in the column (Meikap and Biswas, 2004) and that smaller bubble size leaded to higher collection efficiency of nanoparticles (Hermeling and Weber, 2010). The capture mechanisms that take place in a bubble column depend mostly on the turbulence intensity in the system. To this end, the proposed design was a bed of a glass beads to favour bubbles break up and recombination and turbulence increase. This unit is called "three-phase bubbles column". Chervet et al., (2011) reported that for particles between 20 and 40 nm the removal efficiency of a bubble column dropped off. In order to avoid this efficiency reduction, the three-phase bed column was assisted by a particle enlargement process based on the heterogeneous condensation phenomena. The heterogeneous condensation is important in the fields of meteorology, cloud physics (Hegg, 1990), aerosol science and measurement techniques (Hering et al., 2007)). This technique consists of the vapour condensation on the ultrafine particles when the vapour saturation level is above 1. In this way, a coarser liquid–solid aerosol is generated, making possible the use of conventional technologies for particle capture.

For example, Heidenreich (1995) reported that a cyclone preceded by a heterogeneous condensation process improved its performances.

This work resumes the preliminary results on the removal efficiency of ultrafine polystyrene particles by means a system based on the three-phase bed column assisted by the heterogeneous condensation process. Experiments included the measurement of particles enlargement as a function of the water vapour supersaturation level and the corresponding values of the particles removal efficiency achieved in the three-phase bubble column.

2. Materials and methods

The target particles for this study were calibrated nanoparticles, provided by the Thermo-scientific (Opti-Bind 0.2) as a suspended solution of 15 mL of water with a particles concentration of 10 % in weight, with an addition of sodium azide as surfactant in the order of 0.05 %. The volumetric mean diameter (VMD) was 200 nm.

The experiments were carried out in a lab-scale plant, showed in Figure 1, composed by an aerosol generator, a growth tube and a three-phase-bed column. Pictures of the growth-tube and the three-phase bed column are shown in Figure 2.

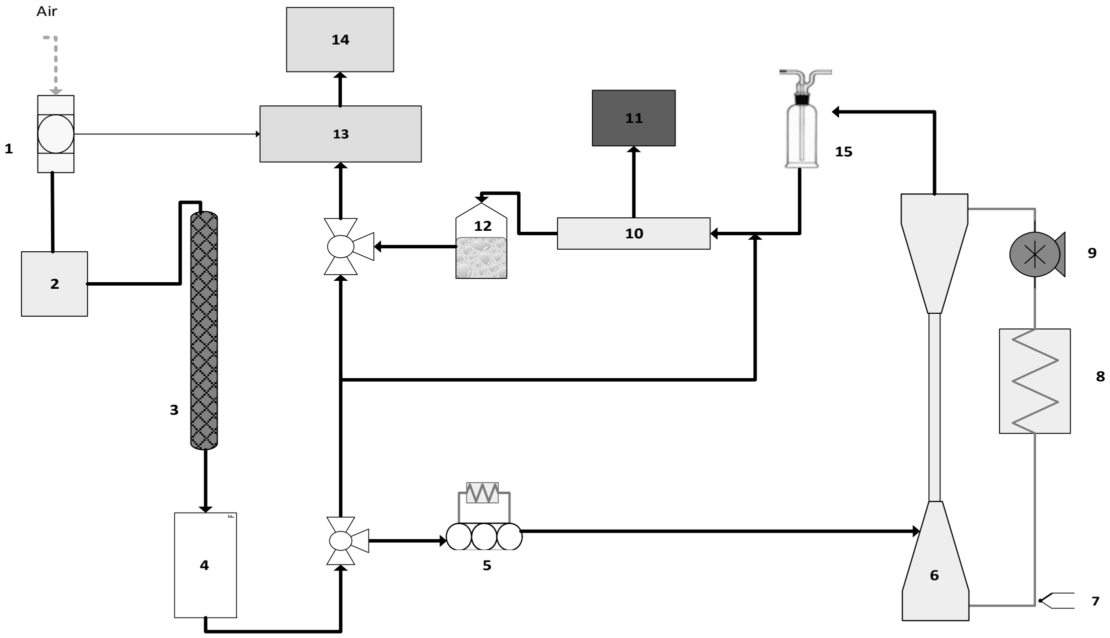

Figure 1: Experimental set up:1-Hepa filter;2-Topas ATM221 aerosol generator;3-Diffusion dryer;4-Flowmeter;5-Saturator;6-Growth tube;7- Thermocouple;8-Thermostatic bath;9- Verder gear pump;10-Quite tube;11- Laser Aerosol spectrometer TSI 3340;12- Three phase bubble column; 13; Dilutor Palas KHG 10; 14-SMPS TSI Nanoscan 3910, 15-Drechsel

The gas with a flow rate of 48 L/h was produced by an aerosol generator TOPAS ATM 221 (2) equipped with a Laskin nozzle and fed with a solution containing 50 mL of bidistalled water and 2 mL of solution of polystyrene calibrated suspended nanoparticles. Before usage, the solution was kept for 10 min in an ultrasound bath at 80 kHz to optimize the nanoparticles dispersion. The particle-laden gas passed through a diffusion dryer (3) to remove water, producing a dry aerosol, and then entered to the growth tube (6), prior saturation in a bubble saturator (5) at 25 °C. The growth tube was the enlargement particle unit where the heterogeneous condensation of the vapour occurred. It consisted in a glass cylinder with length of 40 cm and internal diameter of 1.5 cm. The tube size was determined taking in account the diffusion rate of vapour from the walls to centreline and the need of minimizing the interferences between the gas and the liquid flows. The liquid flow was fed to a bowl placed at the top of the glass and, once it was filled to the brim, a water film flowing along the tube wall was generated, (Figure. 2). A gear pump (9) guaranteed the uniformity of water circulation. The aerosol after the growth-tube was fed to a drechsel (15) in order to remove the water condense. The three way valves allowed to measure the aerosol inlet distribution and particle concentration by the monitoring systems.

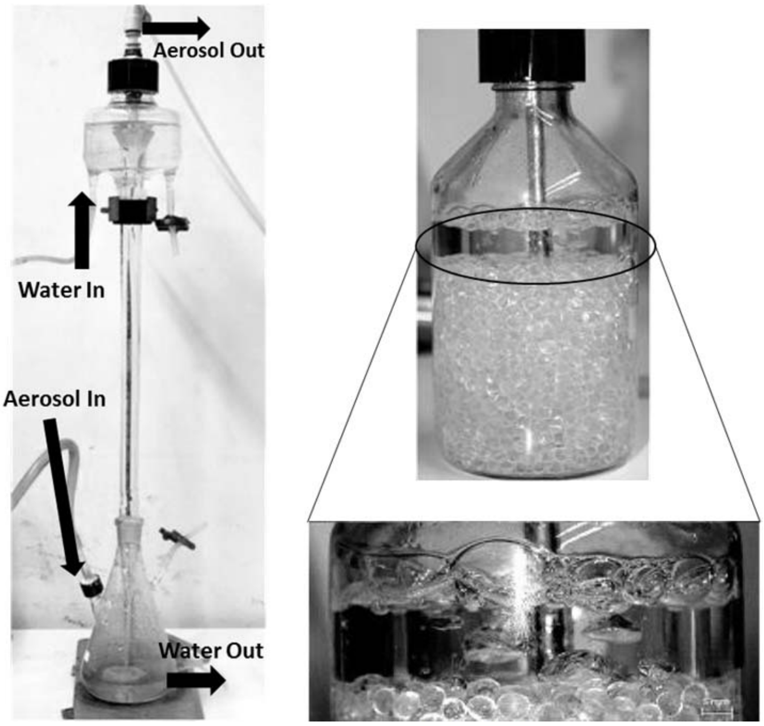

Figure 1: Details of the Growthtube on the left and the three-phase bubble column on the right

In the growth-tube, vapour condensation occurred until the temperature of the gas was lower than that of the liquid film. To this aim, the liquid film temperature was kept at a desired value, T_w, ranging from 30 °C to 80 °C by means of a thermostatic bath (8), and liquid temperatures at the bottom of the growth tube was measured. The film temperature and the gas velocity controlled the supersaturation level (Tammaro,et al. 2012)

The three-phase bubble column (12), showed in Figure 2, was the removal unit where the gas bubbled through a liquid. It consisted in a glass column filled up with 5 mm glass beads and water, kept at room temperature. The total bed height was 9 cm and raised up to 10 cm when the gas was fed. The gas bubbles generation was accomplished by a porous disk placed 8 cm below the liquid level.

Two sets of experiments were performed by varying the film liquid temperature. The first set measured the particle enlargement in the growth-tube and was carried out according to Tammaro et al (2012). The second set measured the particle removal efficiency of the three-phase bubble column assisted by the heterogeneous condensation.

In order to measure the nanoparticles growth, the liquid-solid aerosol was sampled at the growth-tube exit and the particle size distributions were monitored using the Laser Aerosol Spectometer TSI 3340 (11) that is able to measure both liquid and solid with particle size between 90 nm and 7.5 μm, with a maximum particles flow rate of 3,000 particles/s.

Downstream the removal unit, the aerosol size distribution (ASD) and the total number concentration were measured using a scanning mobility particle size (SMPS TSI Model 3910) (14) that allowed measuring solid particles in the range 11.5 – 350 nm. Before entering SMPS, the gas stream passed through a dilution system in order to decrease the particle concentration and to remove water droplets. The dilution system (13) (PALAS KHG 10) was a perfect mixed chamber where a filtered air stream is mixed with the aerosol and a dilution ratio of 10 was set up. In order to remove any liquid in, the chamber was kept at 120 °C. Sampling points were placed at the growth tube inlet and at three-phase bed outlet. The total removal efficiency was evaluated as following:

$$\eta_{tot} = \frac{N_{in}^{tot} - N_{out}^{tot}}{N_{in}^{tot}} \qquad (1)$$

where N_{in}^{tot} and N_{out}^{tot} were, respectively, the particle number concentrations at growth tube inlet and at three-phase bed column outlet. Beyond the total efficiency, it was possible to evaluate an efficiency for the i^{th} particle size channel, with a particle number concentration of N, 1/cm^3 as:

$$\eta = \frac{N_{in} - N_{out}}{N_{in}} \qquad (2)$$

All experiments were repeated in triplicate and the standard deviation was lower than 12 %.

3. Experimental results

The aerosol size distributions (ASD) measured by both the TSI 3910 and the LAS 3340 instruments, at the growth tube inlet, are in Figure 3. It is worth noticing that the two instruments are not easily comparable; they sweep a different particle size interval with a different number of detection channels; in addition, LAS 3340 measures an optical diameters deriving from light scattering, while TSI 3910 measures electrical mobility diameters.

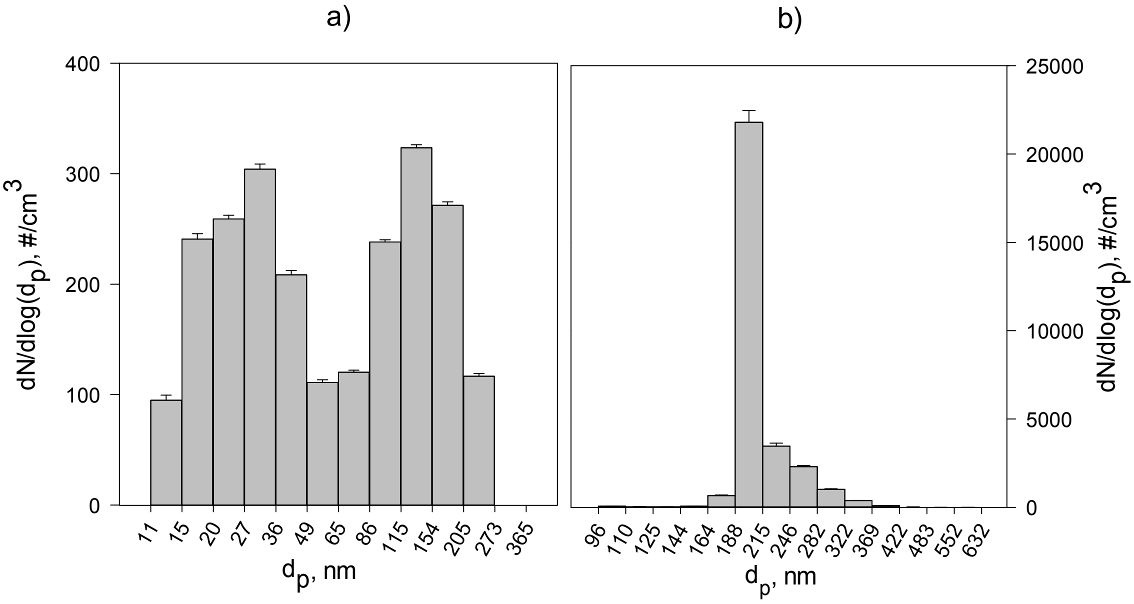

Figure 3: OptiBind Aerosol size distributions at growth-tube inlet by: a) TSI Nanoscan 3910; b) TSI LAS 3340

The OptiBind ASD, monitored by the TSI 3910, showed a bimodal distribution, with particle diameters ranging from 11 to 273 nm; the first peak was at 27 nm and the second one at 115.5 nm. According to the particle manufacturer, the first peak was an experimental artefact deriving from the drying of the surfactant used in the particle solution to avoid coagulation phenomena, so only the second peak was effectively related to the polystyrene nanoparticles. The Laser Aerosol spectrometer revealed a particle narrow distribution, with a mean volumetric diameter around 200 nm. The data errors were always less than 5 %.

The particle enlargement in the growth-tube, at different water film temperatures, are showed in the Figure 4. We preferred to show the particle cumulative distribution function (CDF), instead of the particle size distribution, since it was the easiest and clearest way to visualize the nanoparticles growth. CDFs shifted toward the right side of the graph while water film temperature increases. This result revealed the generation of larger particles, due to the heterogeneous condensation (Tammaro et al. 2012). The OptiBind aerosol had a clear right shift at 70 and 80 °C, therefore the polystyrene nanoparticles triggered an intense heterogeneous condensation at these temperatures. However, a measurable increase of particles at 50 and 60 °C took place, and particle finer than 100 nm were enlarged forming an aerosols with size larger than 100 nm. It is worth noticing that, due to its internal calibration, the LAS 3340 provided a conservative estimation of particles enlargement, because of the lower refractive index of the water than that one of particles with which the instrument was calibrated.

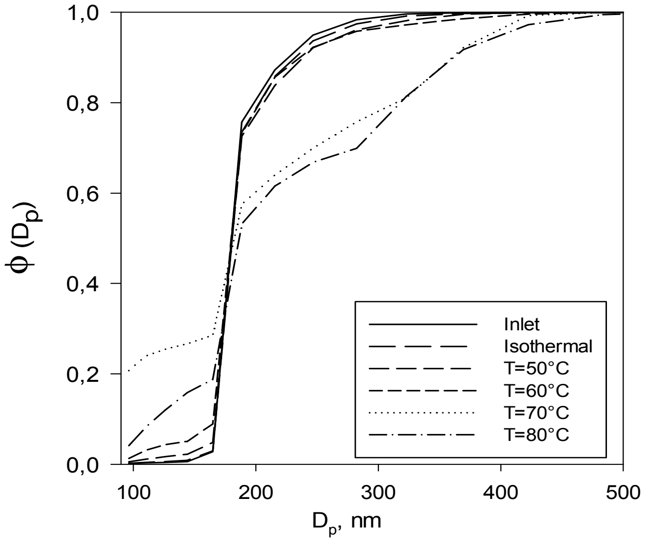

Figure 4: Cumulative distribution functions of the OptiBind aerosol at different water film temperature

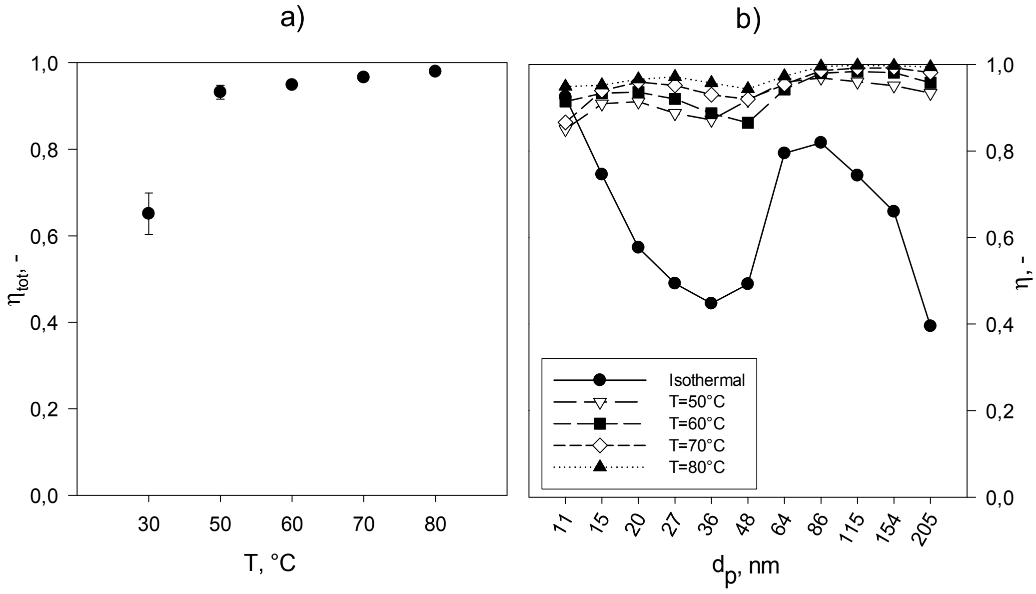

Figure 2: Total particle removal efficiency (a) and removal efficiency with the particle size (b) as function of the water film temperature in the growth-tube

The nanoparticles abatement was studied by evaluating the total particle removal efficiency and the removal efficiency for each particle size at different water film temperatures. Results are shown in the Figure 5. In particular, Figure 5(b) shows that, for the isothermal test, by increasing the particle size, the removal efficiency first decreased, according to the reduction of the Brownian collection mechanism, then it increased up to reach a maximum around 80 nm. This "unusual" non-monotonic trend was already observed with bubble columns by Charvet (2011), but a definitive explanation was not assessed. The non-monotonic trend almost disappeared when the particles were enlarged by passing through the growth-tube; and for water film temperatures ≥ 50 °C the total removal efficiency became higher than 90 % (Figure 5(a)). A possible explanation for these findings is that larger particles had larger chances to be captured by turbulence effects, as vortex shredding and small scale inertial effects. Moreover, the particles with a water shell could be easily captured in the three-phase bed column due to a stronger interface affinity.

4. Conclusions

A new ultrafine particles removal system was presented. The system consisted in the sequence of a growth tube and a three-phase bubble column. First, nanoparticles of polystyrene were tested to study the heterogeneous condensation of water vapour in the growth-tube, set at different temperatures to obtain different super-saturation levels. The nanoparticle presented a significant growth at 70 °C. The particle removal efficiency in the three-phase bubble column was then evaluated. Results show that the enlargement of particles, obtained with the heterogeneous condensation, increments the removal efficiency that results equal to 90 % at water film temperature of 50 °C and even 99 % at water film temperature of 80 °C Thanks to the promising results obtained in these experimental tests, the presented system earns a chance to become a reliable alternative aerosol treatment for industrial application.

References

Charvet A., Bardin-Monnier N.,Thomas D., 2011, Can bubble columns be an alternative to fibrous filters for nanoparticles collection?, Journal of Hazardous Materials, 195, 432–439, DOI:10.1016/j.jhazmat.2011.08.064.

D'Addio L., Di Natale F., Carotenuto C., Balachandran W., Lancia A., 2013, A Lab-scale System to Study Submicron Particles Removal in Wet Electrostatic Scrubbers, Chemical Engineering Science, 97, 176-185, DOI:10.1016/j.ces.2013.04.006.

Di Natale F., Carotenuto C., D'Addio L., Lancia A., Antes T., Szudyga M., Jaworek A., Abbod M., Balachandran W., 2013, New Technologies for Marine Diesel Engine Emission Control, Chemical Engineering Transactions, 32, 361- 366,DOI:10.3303/CET1332061.

Di Natale F., Carotenuto C., 2015, Particulate matter in marine diesel engines exhausts: Emissions and control strategies, Transportation Research Part D: Transport and Environment, 40, 166-191, DOI:10.1016/j.trd.2015.08.011.

Di Natale F., Carotenuto C., D'Addio L., Jaworek A., Krupa A., Szudyga M., Lancia A., 2015 Capture of fine and ultrafine particles in a wet electrostatic scrubber, Journal of Enviromental Chemical Engineering, 3, 349-356, DOI:10.1016/j.jece.2014.11.007.

Hegg D.A., 1990, Heterogeneous production of cloud condensation nuclei in the marine atmosphere, Geophysical research letters, 17, 2165-2168, DOI:10.1029/GL017i012p02165.

Heidenreich S., Ebert F., 1995, Condensational droplet growth as a preconditioning technique for the separation of submicron particles from gases, Chemical Engineering and Processing: Process Intensification, 34(3), 235–244, DOI:10.1016/0255-2701(94)04009-5.

Hering S.V., Stolzenburg M.R., Quant F.R., Oberreit D.R., Keady P.B., 2007, Aerosol Science and Technology, 39, 659-672, DOI:10.1080/02786820500182123.

Hinds W.C., 2011 Aerosol technology: properties, behavior and measurement of airborne particles, 2nd edition, Wiley, New York.

Kantarci N., Borak F., Ulgen K.O., 2005, Bubble column reactors, Process biochemistry, 40, 2263-2283, DOI:10.1016/j.procbio.2004.10.004.

Kerr R.A., 2013, Soot is warming the world even more than thought, Science, 339(6118),382, DOI:10.1126/science.339.6118.382.

Meikap B., and Biswas M., 2004, Fly-ash removal efficiency in a modified multi-stage bubble column scrubber. Separation and Purification Technology, 36(3), 177–190, DOI:10.1016/S1383-5866(03)00213-2.

Menon S., Hansen J., Nazarenko L., Luo Y., 2002. Climate effects of black carbon aerosols in China and India, Science, 297(5590), 2250–2253, DOI:10.1126/science.1075159.

Pope C.A.,Burnett R.T., Thun M.J., Calle E.E., Krewski D., Ito K, Thurston G.D., 2002, Lung cancer, cardiopulmonary mortality, and long-term exposure to fine particulate air pollution, The Journal of the American Medical Association, 287(9),1132–1141, DOI:10.1001/jama.287.9.1132.

Seinfeld, J.H., Pandis, S.N., 1998. Atmospheric Chemistry and Physics. Wiley, New York, USA.

Tammaro M., Di Natale F., Salluzzo A., Lancia A., 2012, Heterogeneous condensation of submicron particles in a growth tube, 74, 124-134, Chemical Engineering Science, DOI:10.1016/j.ces.2012.02.023.

Tsai C., Lin C., Wang Y., Hunag C., Li S., Wu Z., Wang F., 2005, An efficient venturi scrubber system to remove submicron particles in exhaust gas, Journal of the Air & Waste Management Association, 55(3), 319-325, DOI:10.1080/10473289.2005.10464622.

Yao H., Hansen M., Feilberg A., 2014, Dms removal in a bubble reactor by using peroxone (o3/h2o2) reactions, Chemical Engineering Transactions, 40, 229-234, DOI:10.3303/CET1440039.