

The heterogeneous condensation. An effective way to adapt conventional dust removal techniques to new environmental challenges

The emission of particulate matter entrained in industrial and vehicle exhaust gases is one of the major health and environmental concerns. Social awareness on the effects of particulate pollution in urban area has increased sensibly in the past few years moving politics toward the introduction of better environmental regulations.

The traditional particle abatement devices are mainly designed and optimized to treat particles with sizes above 1 μm , and they are far less efficient in collecting sub-micrometric particles, especially in the range 0.1-2 μm , called Greenfield gap. The heterogeneous condensation is a promising technique to improve the performances of traditional particle collection devices. This technique consists in condensing vapor on the ultrafine particles in order to create a coarser liquid-solid aerosol larger in size than the upper limit of the Greenfield Gap. This paper reports the results of an experimental activity, which are described by using consolidated models for heterogeneous condensation. A description of the experimental plant specifically designed and built-up, located in the laboratory of UTTP-CHIA in the ENEA Research Centre Portici, is provided

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La condensazione eterogenea. Uno strumento efficace per adeguare le tradizionali tecniche di rimozione del particolato alle nuove sfide in campo ambientale

L'emissione di particolato contenuto nei gas di scarico delle industrie e dei veicoli è uno degli argomenti più attuali per la salute umana e l'ambiente. La consapevolezza sociale sugli effetti negativi derivanti dall'inquinamento da polveri sottili in aree urbane è aumentata sensibilmente negli ultimi anni e questo ha spinto la legislatura verso la definizione di nuovi e più stringenti limiti di emissione. I tradizionali dispositivi di abbattimento delle particelle sono principalmente progettati e ottimizzati per il trattamento di particelle con dimensioni superiori a $1\ \mu\text{m}$, quindi sono meno efficienti nella cattura di particelle submicroniche, soprattutto nel range $0,1\text{-}2\ \mu\text{m}$, chiamato Greenfield gap. La condensazione eterogenea è una promettente tecnica per migliorare le prestazioni dei dispositivi tradizionali di rimozione delle particelle. Questa tecnica consiste nella condensazione del vapore sulle particelle ultra fini, al fine di creare un aerosol liquido-solido la cui dimensione è maggiore del limite superiore del Greenfield gap. Questo lavoro riporta i risultati di un'attività sperimentale, che sono poi descritti utilizzando modelli consolidati per la condensazione eterogenea. Infine è fornita una descrizione dell'impianto sperimentale, appositamente progettato e costruito, che si trova nel laboratorio di UTTP-CHIA nel Centro Ricerche ENEA di Portici

Introduction

The emission of particulate matter entrained in flue gas issuing from industrial and vehicle exhaust systems is one of the major health and environmental concerns [1]. The entrained very fine inhalable particles can remain suspended in the air for a long time, travelling long distances from the emitting sources, and once inhaled, they can reach the deepest regions of the lungs causing respiratory and other diseases, thus causing severe health hazards. These include heart diseases (strokes, high blood pressure, arteriosclerosis, heart attack) and altered lung functions (asthma, difficult or painful breathing, and chronic bronchitis), especially in children and elder people. In particular, fine particulate matter associated with diesel engine exhaust emissions is also recognized as a carcinogenic substance and, therefore, listed as a mobile air toxic source.

The particle size determines the capacity of penetration in the respiratory organs and the point of deposition of substances within the body. Toxicological studies [2] found an association between

exposure to ultrafine particles and mortality due to respiratory or cardiovascular disease. In fact, particles finer than 100 nm are capable of penetrating the cellular membrane, where they create an inflammatory response by means of oxidative stress, and cause damages to the mitochondria. Particulate emission sources are mainly related to industrial activities, even though the major exposure risks are related to those sources active in urban areas - such as domestic heating, urban traffic and emission from the diesel engines of harboured vessels - usually accounting for 30% of the total particulate emission inventories. Furthermore, nanoparticles are also formed in the atmosphere by nucleation events related to photochemical processes. These nanoparticles are important to cloud formation, but they can also be transported in the atmosphere over large distances and can eventually give rise to additional human exposure via inhalation routes. Nanoparticles can also be deposited onto the soil and water bodies and result in secondary contamination or other environmental effects. Even though the toxicity of particulate matter is

TABLE 1 Treatment techniques for particulate abatement from gas streams

Device	Size (μm) (Efficiency)	Advantages	Disadvantages
Electrostatic Precipitators (dry and wet)	1-2 (95-98%) 0.5 (91%) 0.1 (95%)	<ul style="list-style-type: none"> • High efficiency even for small particles. • Suitable for wide temperature, pressure and gas flow ranges. • Low pressure drop, hence the energy required tends to be low. 	<ul style="list-style-type: none"> • Explosion risk with dry ESP. • The separation capacity depends on the resistivity of dust particles. • Dry ESP not recommended to remove sticky or moist particles.
Wet scrubber (Spray Towers)	1-2 (70-85%) 0.5 (60%) 0.1 (93%)	<ul style="list-style-type: none"> • Flammable and explosive dusts can be handled with little risk. • Simultaneous removal of dust and inorganic compounds. 	<ul style="list-style-type: none"> • Relatively low mass-transfer efficiencies. • Relatively inefficient at removing fine PM.
Fabric Filter	1-2 (99.7-99.8%) 0.5 (99.1%) 0.1 (99.8%)	<ul style="list-style-type: none"> • High collection efficiencies for coarse and fine particles. • Residual emissions are virtually independent of the intake concentration. 	<ul style="list-style-type: none"> • No wet or sticky dusts allowed as input. • There is an explosion risk.
High Efficiency Cyclone	1-2 (20-40%) 0.5 (8%) 0.1(-)	Simple plant and low cost.	For lower diameter it needs to be enhanced by another method.

Source: elaborated by the authors

well recognized since the early years of the industrial era, a significant portion of the existing knowledge on particulate matter toxicity and on the diagnostic methods to determine particle size and concentrations in gas streams have advanced significantly during the past years.

As a direct consequence environmental regulations have gradually reduced the minimum, “cut-off”, particle size allowed at the emission point of industrial exhaust systems from 10 μm (PM10) to 1 μm (PM1). Similarly, the adoption of restrictive regulations is envisaged for diesel engines (e.g., Euro 5 and 6 regulations for cars; USA Tiers 2 and 3 standards for diesel locomotives). These stricter emission limits need engineering challenges both for defining very low particulate emission processes, and the implementation of appropriate filtering systems able to pull down the fine particles [3-4]. However, despite this scenario, the conventional particulate removal devices are generally optimized for particles larger than 1-2 μm and they are far less efficient in collecting submicron particles, especially in the range 0.1-2 μm , called *Greenfield gap*. Usually, complex systems are employed, including trains of consecutive, different, abatement

units (cyclones, fabric filters; electrostatic precipitators; wet scrubber). In Table 1 a brief description of the main traditional particle abatement device is reported.

The enlargement of the particle size through condensational growth is one of the oldest approaches for aerosol measurement and many particle counters are based on this principle [5-6]. Subsequently, condensational growth has been used to enable the collection of particles for chemical analysis or to permit aerodynamic focusing and concentration of ultrafine particles. Finally, water vapour heterogeneous condensation has been exploited as an efficient technique for enlarging particles’ diameter prior to conventional separators [7-8]. Particles undergoing water vapour heterogeneous condensation increase their diameter through a deposition of a water film upon the external surface (Figure 1). Once the droplet with inclusions raises the size over the Greenfield gap, these can be effectively removed by conventional removal devices. In this paper the heterogeneous condensation as a method for growing sub-micrometric particles by means of water vapour condensation – so the grown particles can be separated by a traditional

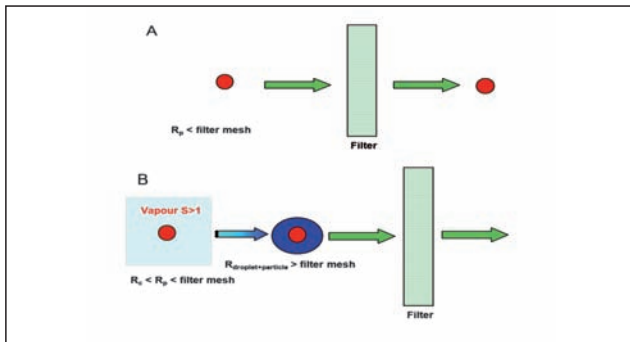


FIGURE 1 Scheme of a mechanism to remove particle from gas without (A) and with (B) heterogeneous condensation, through a conventional device used as a filter
Source: Tammaro [14]

solid-gas separation treatment – was studied. For this purpose, specific equipment has been designed, realized and tested. This experimental plant allows the heterogeneous condensation of water onto the sub-micrometric particles coming from the combustion smokes. Experiments can be proficiently described by using consolidated models for heterogeneous condensation.

Theoretical framework

The Kelvin's equation

Heterogeneous condensation of vapour on a particle is an energetically unfavourable process because the increase in liquid free surface causes a rise in the free energy [5-6]. In order to overcome the energetic barrier and to activate condensational growth, the vapour must be oversaturated. That is, the ratio S (known as *supersaturation*), between partial vapour pressure P_v and equilibrium vapour pressure, $P^o(T)$, at the flow gas temperature T , must exceed the unit [5-6-9]:

$$S = P_v / P^o(T) \quad (1)$$

The smaller the particle the higher the supersaturation required to activate condensational growth. This is because the equilibrium vapour pressure over a droplet is higher than that over a flat surface as a result of the curvature effects on the droplet surface tension [5]. This effect is described by the Kelvin's relation, which associates the equilibrium vapour supersaturation with the diameter of a

droplet composed of that condensed vapour, d_{cr} , called *critical diameter*,

$$d_{cr} = \frac{4\sigma M_w}{\rho R_v T \ln S} \quad (2)$$

where M_w , ρ , and σ are the molecular weight, liquid density, and surface tension of the condensing species, respectively; R_v is the universal gas constant.

The d_{cr} is a property of the condensing species and is equal to the diameter of a droplet in equilibrium with its vapour at S and T . In the case of heterogeneous condensation, the value d_{cr} represent the theoretical minimum size of the solid particle necessary, given S , to activate the condensation process onto its own surface (i.e., the *activation diameter*) [5-6].

Nucleation and growth to droplet

The process of particle enlargement by heterogeneous condensation can be divided into two steps. The first step is called *nucleation*, that involves the formation of a liquid embryo on the particle surface, and the second one is the *growth*, with a formation of a droplet around the particle through condensation of vapour [10]. The physical and chemical properties of particles (compositions, surface characteristics and wettability) affect heterogeneous characteristics considerably. However, the theoretical model considers spherical, smooth and homogeneous particles of coal.

Nucleation rate modelling

The nucleation rate represents the number of critical embryos created on a particle surface per second. Basically, the nucleation rate increases with the supersaturation of the water vapour and the size of the particle [11]. The Fletcher's model is the first and more important model for the nucleation rate. More recent models use the Fletcher's model for their system, upgraded with experimental data [10]. The rate of formation of embryos of size d_e on a particle of size d_p is given by the equation [10,11,12]

$$J = 4\pi K R_p^2 \exp\left(-\frac{\Delta G^*}{kT}\right) \quad (3)$$

where ΔG^* is the free energy of formation of the

embryo. The ΔG^* can be expressed as:

$$\Delta G^* = \frac{8\pi M_w^2 \sigma^3}{3(R_v T \rho \ln S)^2} f(m, x) \quad (4)$$

where $f(m, x)$ is a “geometrical factor” [11], where x is a dimensionless factor defined as ratio between particle and embryo diameters, and m is defined by:

$$m = \cos\theta = \frac{\sigma_{vs} - \sigma_{ls}}{\sigma_{vl}} \quad (5)$$

with

- σ_{vs} : surface tension between vapour and solid;
- σ_{ls} : surface tension between liquid and solid;
- σ_{vl} : surface tension between vapour and liquid coinciding;
- θ : contact angle between the particle surface and embryo.

Eq. n. 5 is the Young’s equation and is the usual definition of the contact angle θ , provided $-1 \leq m \leq 1$.

Finally, in eq. 3, the value of K is somewhat uncertain and depends in detail upon the nucleation situation. Fletcher [11] adopts for K the value of 10^{25} (1/cm²sec).

Commonly, a critical supersaturation, S_{cr} , is conventionally defined in the pertinent literature [9,10,12] as the level of S required to allow the formation of one liquid embryo of size d_e per second. Application of the Fletcher model for an insoluble and

spherical particle for $J = 1$ (1/s) gives:

$$S_{cr} = \exp \left[\frac{1}{R_v T \rho_l} \sqrt{\frac{8\pi M_w^2 \sigma^3}{3k_B T \ln(\pi K d_p^2)}} f(m, x) \right] \quad (6)$$

The S_{cr} is strongly affected by the particle size. In particular it decreases as the particle diameter increases. Then a particle can be activated at saturation equal or higher than its critical saturation.

Growth modelling

Once the liquid embryo becomes stable on the particle surface, a liquid-solid aerosol is formed and the liquid embryo can enlarge thanks to vapour condensation that is regulated by the classical laws of heat and mass transfer. In spite of the geometrical differences, the embryo is usually modelled as a single spherical droplet. Heidenreich [8], in a review on the condensational droplet growth in the air-water system, demonstrated that heat flux between droplets and vapour can be related only to heat conduction and by diffusing molecules, neglecting the Dufour effect. The same author showed that the mass balance on a droplet in a binary air-water gas mixture can be taken into account only for the mass flux due to diffusion: the Stefan-flow and the Soret effect contributions can be neglected since they account for less than 1.5% of the overall mass transfer rate. Under these assumptions the heat and mass balance become [7, 8]:

$$cm_d \frac{dT_d}{dt} + h_l I = Q \quad (7)$$

$$\frac{dm_d}{dt} = \frac{1}{2} \pi \rho_l d^2 \frac{dd}{dt} = -I \quad (8)$$

$$\text{with } t=0, m = m_e = \frac{\pi d_e^3}{6} \rho_w \text{ and } T=T_e$$

where Q is the total heat flux and I is the total mass flux to the droplet; T_d is the temperature of the droplet and h_l , c , m_d , d are the specific enthalpy of the liquid, the specific heat capacity, the mass and

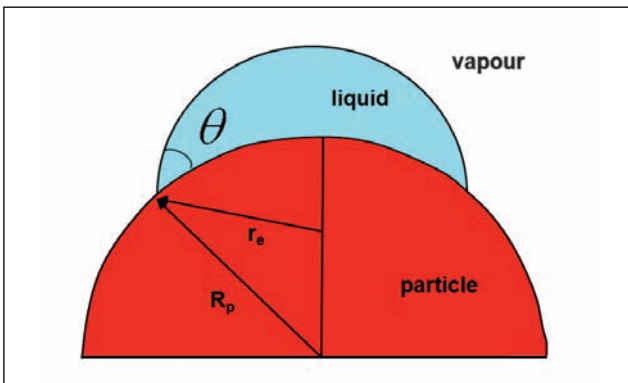


FIGURE 2 Schematic representation of a liquid on a spherical particle (R_p); r_e , radius of embryo; θ : contact angle
Source: Fletcher [11]

the diameter of the droplet respectively; m_e and T_e are the mass and the temperature of the embryo respectively. The expression of I was first given by Maxwell (see Fuchs [13]):

$$I = \frac{2\pi d \cdot M_w D}{R_v T} (P_{v,a} - P_{v,\infty}) \quad (9)$$

where D is the diffusivity of water, T is the film temperature at the droplet surface and $P_{v,a}$ is the vapor pressure at the droplet surface, obtained by the Kelvin's relation.

Through equations n. 8 and 9 it's possible to describe the final diameter of droplet obtained from condensational growth [7,8,14], if the initial embryo size, the properties of the particle surface and the properties of the gas phase are known. In particular, it should be noticed that the proposed equations for heterogeneous condensation can be safely used only if θ is known *a priori*, from specific experiments, or if it is used as a parameter for numerical fitting of experiment [15].

Experimental apparatus

In Figure 3 the experimental system adopted for the heterogeneous condensation tests is described. The particles were produced by premixed ethylene-air flame whose C/O equivalent ratio is adjusted with the two flowmeters (FC1) that controlled the ethylene and air flows to the burner. The flue gases emitted by the flame are diluted with indoor air (the laboratory air was conditioned at 20°C and

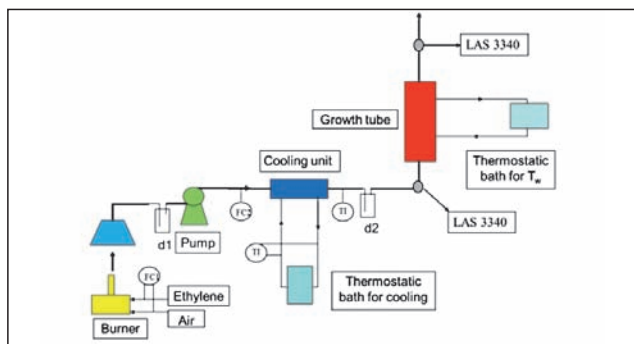


FIGURE 3 Experimental apparatus
Source: Tammaro et al. [15]

30% *circa* of relative humidity) and sampled by means of a hood connected to an extractor fan. The gas flow rate was controlled by the flowmeter FC2. The gas was then cooled to its dew point temperature, i.e., to its saturation level, determined by the temperature measurement. The latter was estimated on the basis of the reaction stoichiometry and the water content in the indoor air. The gas cooling unit was a typical glass condenser with cold water coils inside. Water temperature was controlled by a thermostatic bath. To prevent the presence of accidental droplets in the gas, two Drechsel bottles (d1 and d2) were placed along the gas line before the growth tube. The saturated gas was then sent to the *growth tube* (see following). The aerosol size distribution (ASD) and concentration in gas streams were measured by using a Laser Aerosol Spectrometer (TSI Model 3340) that allows measuring particle size in the range 90-7500 nm.

Growth tube

The most important part of the experimental plant is the one where the so-called *growth tube* – the size amplification of particles' heterogeneous condensation – is realized.

The growth tube is a glass pipe with $L = 40$ cm and $ID = 1.5$ cm. The dimensions of the tube have been specifically fitted for having a permanence time (t_{res}) of the smokes inside the tube higher or equal than $t_{het,cond}$, ($t_{res} \geq t_{het,cond}$), defined as the time needed for the heterogeneous condensation and the growth to occur, which has been assessed as 1 sec [5,6,9,10]. Actually, once fixed t_{res} , the length and internal diameter of the growth tube take into account the thermal and mass diffusivity of air and vapour respectively. Water vapour supersaturation is achieved by introducing an air flow into the growth tube, where temperature is lower than that of the growth tube walls, as illustrated in Figure 4. The walls are actively wetted to maintain a partial pressure of water vapour at the walls near the equilibrium vapour pressure at the wall temperature. The liquid film, on the growth tube wall, has been obtained with tangential inlet of the water. In Figure 5 a functioning scheme of growth tube is shown, where the nucleation and condensational growth steps are highlighted.

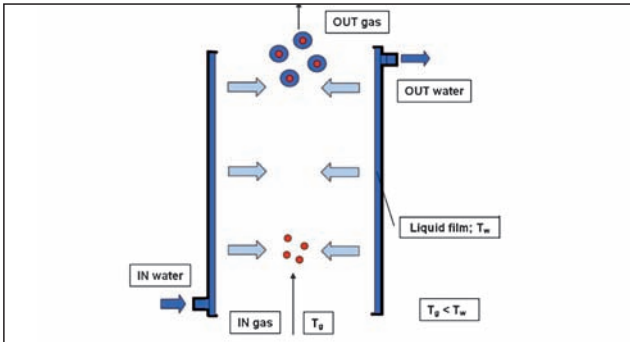


FIGURE 4 Schematic of the growth tube, showing a wet-wall tube, the walls of which are warm with respect to the entering flow. T_w : wall liquid temperature, T_g : gas temperature
Source: Tammaro [14]

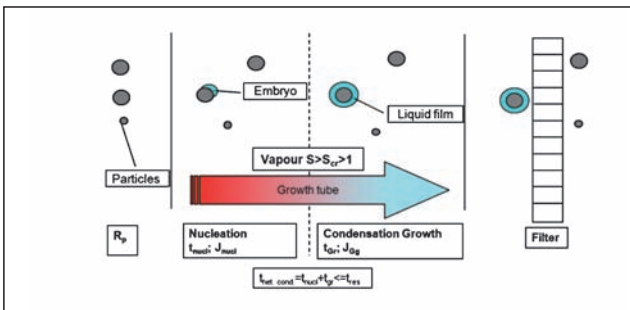


FIGURE 5 Functioning scheme of growth tube with nucleation and condensational growth steps
Source: Tammaro [14]

Experimental Test

In this work the results of an experimental campaign are presented, composed by thirty-six tests, organized in two main groups, corresponding to two equivalent ratios Φ , of 2.38 and 3.30. Each group was composed by six sub-groups, each of which made by three tests with constant residence time (t_{res}), dilution ratio (Dr , equal to the ratio between dilution air and combustion smokes) and total flow (Q_g). Each tern of tests in a group consisted of one “blank test” (without any water in the growth tube) and 2 tests with different water temperatures, respectively of 309 and 317 K. Details of the experimental conditions are reported in Table 2.

During a test, the burner was turned on at given ethylene and air flow rates, and the dilution air flow

GROUP 1: $Q_{C_2H_4} = 59 \text{ ml/min}$, $Q_{air'} = 355 \text{ ml/min}$; $\Phi = 2.38$				
Test	T_w , K	t_{res} , s	Dr	Q_g , l/min
1	Blank test 309 317	0,94	9,86	4,50
2				
3				
4	Blank test 309 317	1,06	8,65	4,00
5				
6				
7	Blank test 309 317	1,21	7,45	3,50
8				
9				
10	Blank test 309 317	1,41	6,24	3,00
11				
12				
13	Blank test 309 317	1,70	5,03	2,50
14				
15				
16	Blank test 309 317	2,12	3,83	2,00
17				
18				
GROUP 2: $Q_{C_2H_4} = 82 \text{ ml/min}$, $Q_{air'} = 355 \text{ ml/min}$; $\Phi = 3.30$				
Test	T_w , K	t_{res} , s	Dr	Q_g , l/min
19	Blank test 309 317	0,94	9,32	4,50
20				
21				
22	Blank test 309 317	1,06	8,17	4,00
23				
24				
25	Blank test 309 317	1,21	7,03	3,50
26				
27				
28	Blank test 309 317	1,41	5,88	3,00
29				
30				
31	Blank test 309 317	1,70	4,73	2,50
32				
33				
34	Blank test 309 317	2,12	3,59	2,0
35				
36				

TABLE 2 Experimental plan
Source: Tammaro et al. [15]

rate, the water flow at the growth tube and the exchanger were fixed at the desired levels. The blank tests were performed in order to take into account all the scavenging phenomena occurring in the entire equipment, when heterogeneous condensation is absent. The two tests at different temperature provide information on the actual particle size distribution at the exit of the growth tube in two different operating conditions.

Figure 7 reports the typical experimental results of heterogeneous condensation tests, compared with the initial particles' distribution. In particular, these

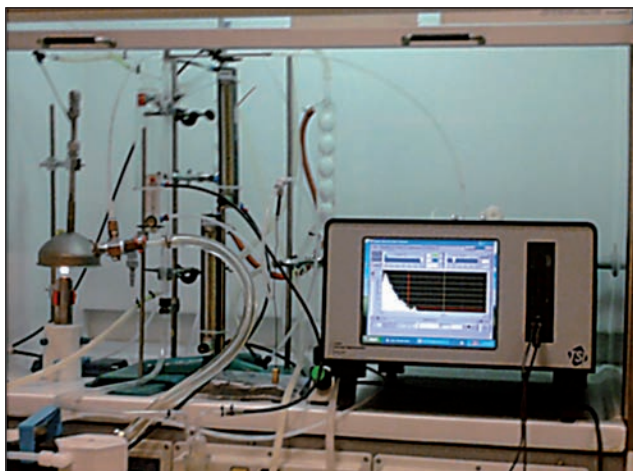
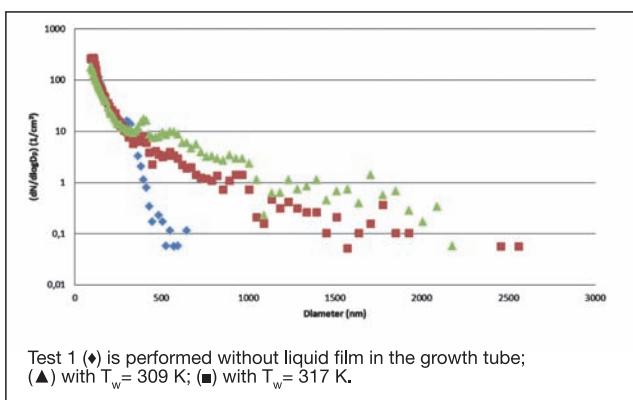


FIGURE 6 Laboratory plant
Source: ENEA



Test 1 (♦) is performed without liquid film in the growth tube; (▲) with $T_w = 309$ K; (■) with $T_w = 317$ K.

FIGURE 7 Size distribution vs diameter in Tests 1-2-3 (Table 2), with $\Phi=2.38$
Source: Tammaro [14]

were obtained for the case of water temperature of 309 and 317 K and residence time of 0.94 s (Test n. 1, 2 and 3 in Table 2).

In Figure 7, the diamond symbols represent the blank tests, i.e., the particle size distribution generated by the flame and measured at the end of the growth tube. The square and triangle symbols represent the particle size distribution at the exit of the growth tube when heterogeneous condensation occurs.

The comparison between the aerosol size distributions with and without condensation clearly points out that below an experimental critical particle diameter (300 nm ca in Figure 7) no condensation occurred. The coarser particles, instead, were enlarged due to the heterogeneous condensation growth and a liquid-solid aerosol with size up to 2.0 μm was formed. All the experimental tests gave results qualitative similar to those reported in Figure 7. Although tests were planned for two different values of Φ , 2.38 and 3.30, no apparent differences were observed from the two cases. The results are quite easy to understand since higher particle residence times means that the particles persist in a supersaturated environment for longer times, allowing the condensation of more vapor and the formation of larger liquid-solid aerosols.

Furthermore, experimental results show that particle enlargement is favored by higher temperatures and, if enlargement of 90 nm particles is desired under the examined conditions, a residence time close to 2 s should be adopted at $T_w = 309$ K, but this time is almost halved at 317 K. Nevertheless, to enlarge all particles above 1 μm and allow their removal by conventional particle collection technologies, longer residence times and higher water temperatures are necessary.

The analysis of the experimental data requires the modelling of the dynamics of embryo nucleation and aerosol growth in the supersaturated environment of the growth tube. The objective of the model is to obtain the size distribution of the liquid-solid aerosol at the exit of the growth tube at the different investigated conditions, once the inlet particle size distribution is known. To this aim, the heterogeneous condensation process was modelled considering the fate of each particle flowing in the growth

tube under the assumptions that:

- the particles were homogeneously distributed in the gas stream;
- the gas flow in the growth tube was laminar and in steady state;
- the particles follow the gas streamlines as they were free of inertia, so that they move longitudinally along the growth tube without changing their radial position;
- the particle concentration was sufficiently low to ensure that heterogeneous condensation negligibly influences the water vapour concentration profile generated by the liquid film in the growth tube.

The first three assumptions are easy to accept once the gas properties and the geometry of the growth tube are known. The last one, instead, has been verified in our experimental conditions [15]. This modelled particle size distribution was compared with the corresponding experimental one and the value of the contact angle, θ , was then determined by best fitting experimental results [15]. Two sample results are reported in Figure 8, which describes the comparison between modelled and experimental values of the particle size distribution at the exit of the growth tube for two different operating conditions (corresponding to tests n. 11 and 3 of Table 2, respectively). Experimental values of the corre-

sponding inlet particle size distributions are also reported. It clearly appears that, at both temperatures, the modelled aerosol size distribution gives a very good representation of experiments. Initial particles' size distributions are also reported.

Conclusions

This work reports the study of the heterogeneous condensation as a technique to pre-conditioning the sub-micrometric particles contained in a gas, with the aim of increasing their dimensions enough to allow the use of consolidate gas cleaning techniques. This problem is particularly important for particles in the Greenfield gap and for ultrafine particles, for which the conventional depuration techniques are quite ineffective.

For this purpose, an instrumented lab scale equipment was designed, constructed and tested. The core of the equipment was the growth tube, which consisted of a glass tube where the particle laden gas flow came into contact with a supersaturated water vapour environment, generated by a liquid film flowing on the tube's internal wall. Experiments showed that by increasing the treatment time, the maximum size of the liquid-solid aerosol increases as well. A descriptive model based on the theories of heterogeneous nucleation and condensational growth was used to estimate the final

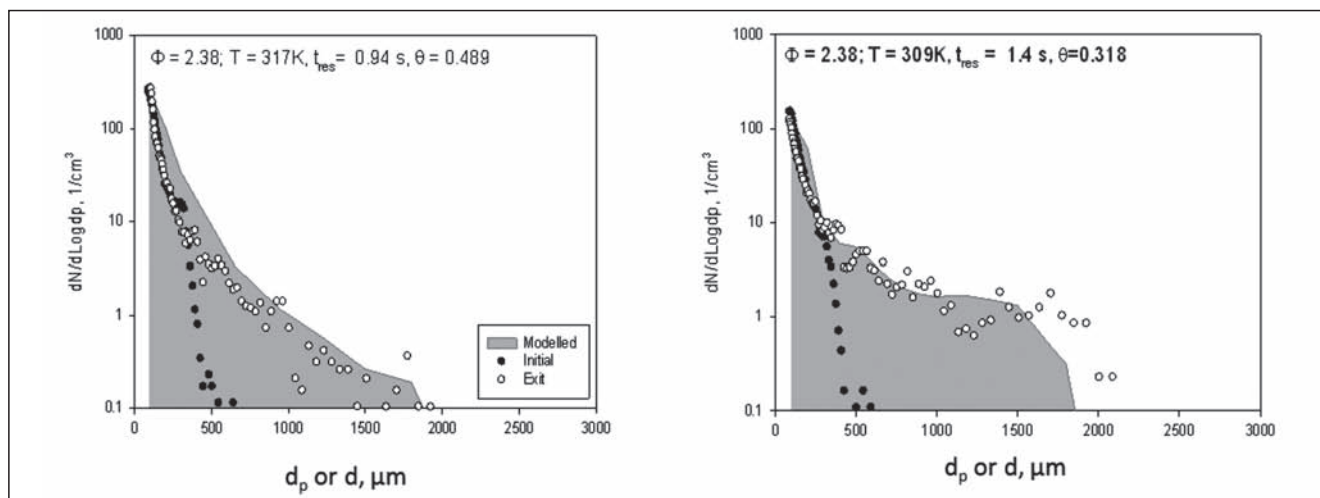


FIGURE 8 Comparison between experimental and modelled particle size distributions at the exit of the growth tube for two different operating conditions
Source: Tammaro [15]



particle size distribution at the exit of the growth tube. The parameter θ , the contact angle, is used here to take into account the discrepancies between the actual experimental conditions and the assumptions at the very basis of the models used to describe the physics of nucleation phenomena, above all the hypothesis of homogeneous spherical particles and of a liquid embryo modelled as a continuous fluid, regardless of the nanometric size of the investigated aerosol.

With reference to the industrial application of growth tube devices, the experimental results showed that the heterogeneous condensation can be easily carried out. Anyway the experimental configuration was not optimizing the heterogeneous condensation and different solutions should be investigated, but for its simple design, this system should be worth considering for large scale industrial applications.

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