HETEROGENEOUS CONDENSATION FOR SUBMICRONIC PARTICLE ABATEMENT: EXPERIMENTS AND MODELLING

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Abstract

This paper reports a study of the heterogeneous condensation of sub-micrometric particles produced by a model ethylene/air flame, performed with an instrumented lab-scale plant. Tests were performed by varying the temperature of the condensing vapour and the gas residence time. Experiments can be described by using consolidated models for heterogeneous condensation, provided that the contact angle is considered as an adjustable parameter, variable with temperature.

Introduction

The emission of particulate matter entrained in industrial and vehicles exhaust gases is one of the major health and environmental concerns (e.g. [1]). Social awareness on the effects of particulate pollution in urban area increases sensibly in the last years, moving politics toward the introduction of more restrictive environmental regulations. In particular, these regulations are gradually reducing the minimum, "cut micrometric particles", especially those in the range 0.1 - 2 μm, called Greenfield gap. Usually, for process industry and combustion units, complex systems including trains of consecutive abatements devices are adopted [2]. These 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 those in the Greenfield gap. Heterogeneous condensation is a promising technique to improve the performances of traditional particle collection devices. This technique consists in the condensation of vapour on the ultrafine particles in order to create a coarser liquid-solid aerosol whose size is larger than the upper limit of the Greenfield Gap. The aim of this work did not aim to proof that heterogeneous condensation is a reliable way to enlarge particle, but was rather focused on establishing how gas-liquid temperature differences and treatment times affect the condensational growth. To this aim, a dedicated experimental campaign was carried out at low temperature of the evaporating liquid and small treatment times, when heterogeneous condensation is not completed for all the particle sizes, and the effect of process parameters can be better revealed. A comparison between experimental results and a model was performed.

Experimental apparatus

Figure 1 describes the experimental plant for the heterogeneous condensation tests. The particles are produced by a premixed ethylene-air flame. The equivalent ratio, Φ, of the flame is adjusted through two flowmeters (FC1-2) that control the ethylene and air flows to the burner. The gas emitted by the flame are diluted with ambient air and fed, by means of a hood connected to a pump, to a cooling unit. In this unit, the gas sample is cooled to the dew point temperature of the water vapour so that the gas exiting the cooling unit is saturated with water. The gas cooling is achieved by indirect contact with cold water, whose temperature and flow rate are controlled through a thermostatic bath and fixed on the basis of the reaction stoichiometry and the indoor relative humidity (30% *circa* measured by a hygrometer) information. The cold and water saturated gas stream is then sent to the growth tube where the hot water film generate a supersaturated environment and heterogeneous condensation occurs. Finally, the outflowing gases are analysed in terms of size and concentration of particles by using a LAS Model 3340 by TSI, which allows measurement of particle sizes in the range between 90 and 7500 nm. The growth tube consists in a 40 cm length and 1.5 cm internal diameter glass cylinder with entrance and exit connections for both the cooled gas and the water. Gas flow rate (between 2 and 4.5 l/min) and tube length were chosen to operate with a gas residence time, t_{res}, spanning between 0.94-2.2 seconds *circa*, that are comparable with indications of former studies [3-5]. The water inlet to the growth tube is tangential so to assure a perfect adhesion of water film with the tube walls. The liquid film temperature is kept at the desired value, T_w, by means of a thermostatic bath, and the water is injected counter-currently respect to the gas stream. The experimental campaign was composed by eighteen tests, corresponding to equivalent ratios Φ of 2.38. The 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 in 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 1.

Results and Discussion

Figure 2 reports the typical experimental result of heterogeneous condensation tests. In particular, these were obtained for the case of water temperature of 309 K and residence time of 0.94 s (Test n. 1 and 2 in Table 1).

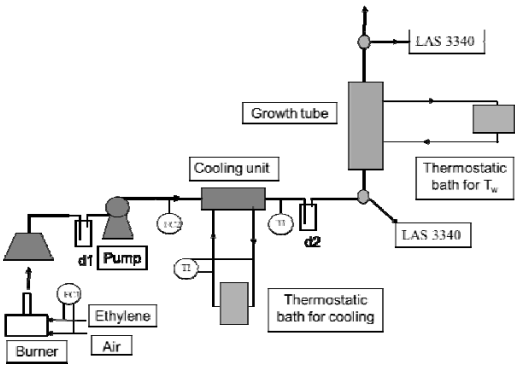

Figure 1. Experimental apparatus. The symbols d1 and d2 indicate Drechsel. FC1 and FC2, indicate flow-meters.

In Figure 2, the full 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 empty symbols represent the particle size distribution at the exit of the growth tube when heterogeneous condensation occurs. The comparison of the two distributions clearly point out that below a critical particle diameter, d_{cr}, no condensation occurred. The coarser particles, instead, were enlarged due to the heterogeneous condensation growth and a liquid-solid aerosol with size up to 2.5 μm was formed. In particular, for the sake of simplicity, we can define a reference maximum particle size, d_{max}, as the larger aerosol size with a given concentration $N(d_{max})$. This concentration is arbitrarily set to 1 particle per cm^3 as reported in Figure 2. All the experimental tests gave results qualitative similar to those reported in Figure 2. The value of the critical diameter gradually moved towards finer particles for longer residence times. Similarly, the values of d_{max}, present a generally increasing trend by increasing the residence time. Furthermore, experimental results show that particle enlargement is favoured 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. However, the degree of particle enlargement, as resumed by the value of d_{max}, appears to be quite less influenced by T_w. Nevertheless, to enlarge all particles above 1 μm and allow their removal by conventional particle collection technologies, higher residence times and higher water temperatures are necessary. The generation of a liquid-solid aerosol by heterogeneous condensation of a vapour on a particle can be divided into two stages that can be assumed as consecutives (e.g.[6, 7]). The first step involves the *Nucleation*, i.e., the formation of a liquid embryo on the particle surface; the second step is the *Growth*, i.e. condensation of the vapour around the embryo and the consequent enlargement of the liquid-solid aerosol.

Table 1. Experimental plan and results (B.t. stands for Blank test).

Test	T_w (K)	t_{res}	Dr	Q_g, (l/min)	Q_{C2H4} (ml/min)	Φ	d_{cr} (nm)	d_{max} (nm)
1	B.t.							
2	309	0.94	9.86	4.50			350	1000
3	317						350	1000
4	B.t.							
5	309	1.05	8.65	4.00			160	1800
6	317						100	1000
7	B.t.							
8	309	1.20	7.45	3.50			160	1800
9	317				59.20	2.38	<90	1100
10	B.t.							
11	309	1.40	6.24	3.00			125	1750
12	317						<90	2000
13	B.t.							
14	309	1.70	5.03	2.50			190	2200
15	317						130	2360
16	B.t.							
17	309	2.10	3.83	2.00			120	2000
18	317						112	2500

In the following, the classical assumptions of smooth, spherical and homogeneous particles are considered. The analysis of the experimental data requires the modelling of the dynamics of embryo nucleation and aerosol growth in 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:

1. The particles were homogenously distributed in the gas stream.
2. The gas flow in the growth tube was laminar and in steady state.
3. The particles follow the gas streamlines like they were free of inertia.
4. The particle concentration was sufficiently low to assure that heterogeneous condensation negligibly influences the water vapour concentration profile generated by the liquid film in the growth tube.

The first three assumptions are easily to accept once the gas properties and the geometry of the growth tube are known. The last one, instead, is been verified in our experimental conditions [8]. 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 of experimental result.

Two sample results are reported in Figure 3, 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, respectively, to the tests n. 11 and 3 of Table 1).

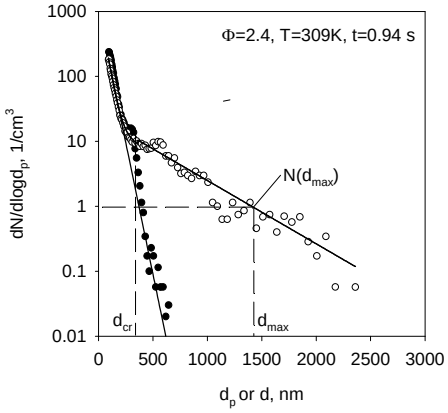

Figure 2. Typical particle size distribution obtained during heterogeneous condensation tests. Full symbols: Blank test n.1; Empty symbols: Test n.2.

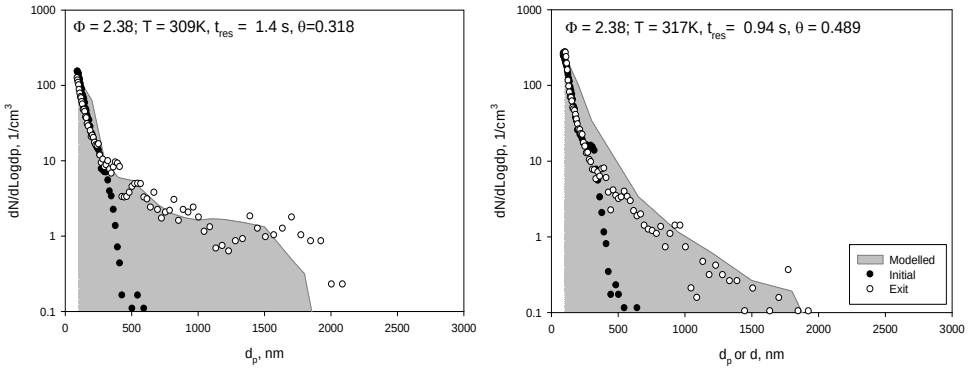

Figure 3. Comparison between experimental and modelled particle size distributions at the exit of the growth tube for two different operating conditions. Particles size distributions at the growth tube inlet are also reported.

At both temperatures, modelled aerosol size distribution gives a very good representation of experiments. In particular best fitting of experimental results was been achieved by using the contact angle of 0.318 rads for T_w=309 K and 0.489 rads for T_w=317 K [8].

Conclusions

This work reported the study of the heterogeneous condensation as a technique to pre-conditioning the sub-micrometric particles contained in a gas with the aim of increase their dimensions enough to allow the use of consolidate gas cleaning

techniques. For this purpose instrumented lab scale equipment was designed, constructed and tested. The core of the equipment was the growth tube, which consisted in 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 internal wall. Experiments showed that by increasing the treatment time, the value of the d_{cr} gradually moves towards finer particles. Similarly, the d_{max}, presented an increasing trend by increasing the residence time. Furthermore, experimental results showed that particle condensation was strongly favored by higher difference of temperatures between liquid and gas. A descriptive model was used to estimate the final particle size distribution at the exit of the growth tube. The results of this model were successfully compared with experimental results, by using a value of the contact angle of 0.318 rads for T_w=309 K and 0.489 rads for T_w=317 K. The parameter θ is used here to take into account for 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 disrespectfully of the nanometric size of the investigated aerosol.

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