Condensation of Inorganic Vapors during Cooling of Synthesis Gas for Gasification of Biomass

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Abstract- **This paper present a study for the condensation of inorganic species in a Biomass to Liquids (BtL) process. The experimental design was based on operating conditions similar to those of a biogas gasification plant mass and physical phenomena that occur during the condensation of aerosols. To obtain conditions simplified but representative of a BtL process, a potassium chloride (KCl) vapor and solid particles of carbon are cooled to different cooling speeds. These particles are then dipped in a dilution probe and the results are then analyzed using a Low-Pressure Electric Impactor (ELPI) and an electric mobility granulometer (SMPS). The results showed the importance of the phenomenon of direct condensation of KCl on the walls. Seeding with Graphite particles reduced the amount of KCl deposited on the walls.**

Keywords: Biomass; aerosol; inorganic; gasification; modeling..

I. INTRODUCTION

Fuel production by the Biomass route to Liquids BtL is a solution to produce liquid fuels using renewable resources. The energy use of biomass can be to do in different ways, combustion, pyrolysis, vapor-gasification, partial combustion ... However, biomass naturally contains inorganic species minority that will be sprayed high temperature, during gasification, for example. Then cooling, the inorganic vapors are going to condense and form particles that are a threat to the process: indeed, these species can corrode reactor walls, butcher hoses, agglomerate fluidized beds or disable catalysts [1-3]. Because of this, it is necessary to remove these particles from the gas effectively in the process. To optimize the cleaning of the gas, it is necessary to know when and how particles are formed. Studies have already been concerning condensation and the deposit of particles in combustion processes the biomass, but inorganic species do not do not involve the same way in burning and gasification. Sulphates, for example, which are predominant in combustion are absent when steam gasification due to the low potential oxygen [4, 5]. Thus, the developed models in the case of biomass burning cannot be used directly in the case of gasification, and new models must be validated. Different studies were performed to study the condensation of inorganic vapors during cooling of synthesis gas for gasification of biomass such as the studies of Samiran et al [6], Madadian et al. [7] and Neves et al. [8]. An experimental device was thus developed to study the condensation of inorganic species under conditions representative of industrial installations and subsequently to validate the models. In this article will be presented results obtained on the device, as well as an application deducted from them in order to reduce inorganic deposits on the walls.

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II. DESCRIPTION OF PHENOMENOLOGY AND MODELS

The phenomenology of condensation

Condensable vapors are condensed in a series of phenomena simultaneously. These phenomena are recalled in Fig. 1. Thus, during the cooling of condensable vapors, with the increase in supersaturating and in the absence of seeding particles, germs will form randomly. These germs are unstable as long as they remain below a critical size.

Once the critical size is exceeded, the nuclei are formed and stable. It's nucleation. Simultaneously, in case of prior presence of seeding particles, the vapors will condense on the particles present by heterogeneous condensation. This phenomenon causes the decrease of vapor in the medium and will compete with the nucleation. In practice, the presence of particles in the flow should be sufficient to prevent homogeneous nucleation. Finally, all the particles present in the flow can interact during the agglomeration phenomenon. This phenomenon does not lead to a variation of the condensed mass but importantly, particle size and particle morphology. The last phenomenon to take into account is the formation of deposits on the walls: they can occur either by direct condensation of the vapors on the walls, or by deposits of the solid particles already formed in the flow on the walls. All of these phenomena are described in individual models and grouped in a model called "global".

Models

The so - called "global" model of aerosol condensation is composed of different "sub - models", each of them describing one of the phenomena that occur simultaneously during a condensation (nucleation, growth, agglomeration, deposits); for each of these individual models, there are several possible modelizations which differ according to the approximations realized. For a particular application, it is therefore necessary to choose a specific model to describe each phenomenon. For the global model, it is possible to start from a model developed and validated for a similar application, for example biomass combustion [9] and modify it to take into account the differences with the application in question, here the gasification of the biomass. This is the solution chosen in this work. Changes to the overall model must naturally be validated by experience. For this purpose, we have carried out condensation experiments of a model aerosol (KCl) in a specific experimental installation. This installation, the experiments carried out and the results obtained are described below. The input / output data of the models to be measured on the experimental set-up are as

follows: temperature profiles, particle size and particle size at the device inlet, KCl partial pressure, number, particle size distribution and particles formed from the aerosol, masses deposited on the walls, mass balances.

III. The Experimental Setup Description

Principle

The ANACONDA device (ANAlysis of CONDensation of Aerosols) was designed to obtain simplified experimental data but representative of the phenomena that occur in an industrial gasification process of biomass (cooling gradients and concentrations solid particles encountered in the installations). It consists in condensing, in a controlled way, condensable vapors of an inorganic species abundant pattern in biomass ash (KCl) in the presence or absence of pre-existing particles simulating soot, for example (graphite), and carry out measures to inform the corresponding physical models.

Running a test Preparation of the gas flow

A suspension of KCl is produced in the form of droplets of water and KCl dissolved by a generator pneumatic, type TOPAS ATM220 or Palas AGF, and injected into a nitrogen stream of 30 NL / min. The concentration of KCl in the generation solution is chosen such that the concentration of KCl in the gas is 4 mg/Nm3 or 40 mg / Nm3. The diameter calculated droplet size is 4.3 μm. The tube run through the flow is long enough (2m) to ensure the drying of the particles before entry into the ovens. This has been verified by particle size measurements of the formed drops. The radius of the tube (1 cm) is chosen to obtain a number Reynolds of 2,500 for a flow rate of 30 NL/min and ensure a good mixture of particles [10]. Particles of graphite are injected at the end of drying thanks to a Palas GFG 1000 graphite generator. The operating parameters of this generator are as follows: 2.5 bar, no dilution, discharge frequency equal to 800 Hz. In order to change the concentration number of carbon particles, part of the outflow of the generator is discharged to outside while the other part is injected into flow. It has been experimentally verified that this division of the flow also led to a division number of particles. Finally, the flow is heated thanks to ovens tubular to reach 800 ° C and allow evaporation KCl.

Cooling

In order to vary the speed cooling, we can change the thickness insulation of the experimental tube in which must be unwind condensation with air knives variables. Three cooling speeds were defined: 300, 500 and 1000 K/s. The length of the tube can be adapted to the chosen speed, but only the cooling rate of 1000 K/s could be tested as part of this work. Temperature outer wall of the tube is recorded by 30 thermocouples fixed on the outer wall. From these measurements is deduced by a simple calculation (1-D, coefficient heat transfer gas / wall) axial profile gas temperature. The length of the cooling tube is 1.3 m and the outlet temperature is of the order of 350 °C.

Tempering and particulate sampling

The particles are removed and analyzed on an impactor low electrical pressure (ELPI) that can be accompanied in parallel with a very high filter efficiency in order to have a measure of the mass of the all the particles that enter the ELPI, and an electric mobility granulometric (SMPS). These measurements (ELPI and SMPS) require a temperature flow of up to 60°C, which makes it necessary to cool the flow without changing the particles formed by air-dilution quenching (dilution factor of 100). This air taken from the air ambient is filtered upstream and downstream of the probe at dilution by THE filters. The flow of the dilution is measured by a diaphragm and regulated by a loss of variable load before the extractor.

IV. Measurements and Experimental Setup

The measurements include the temperature of the outer wall of the section test; the granulometric of the particles leaving the test section after quenching (ELPI and SMPS), The mass of KCl particles collected on each stage of the ELPI, determined by dissolving the deposits of each tray in water then measuring the concentration of K present in the solutions obtained by ion chromatography. He has been verified that the dilution method allowed the total extraction of potassium chloride present on filters and trays. The mass flow of KCl present in the front gas ovens and in the dilution probe and the air flow circulating in the dilution probe. Holman [11] method is used to estimate the uncertainty in experimental results. The minimum error for any instrument is equal to the ratio between its least count and minimum value of the output measured. All values are small compared to the obtained data and found to be within the allowable range of the measurements.

V. Results and Discussions

The first tests were conducted cold and hot in order to check the overall good functioning of the device. 1000 K / s cooling has been imposed for hot testing. The total gas flow in the pipe is 30 LPM. In the flow is injected carbon alone, then KCl alone, and finally KCl and carbon simultaneously. The KCl generator used is Topas ATM220. Granulometric measurements are carried out with ELPI and SMPS. Diameter measurements particles are given here in diameter of mobility equivalent electric. Experimental conditions are summarized in Table 1.

Carbon alone (tests A and B)

Fig. 2 shows the number distributions Carbon particles, without heating (Test A) and after heating (test B). These are almost identical. The electric mobility diameters means are 64.8 nm and 64.2 nm with deviations types of 1.65 and 1.73 respectively. This result, associated with microscopic observation of particles of graphite whose morphology is quite similar between the two tests, leads us to say that the distribution of carbon particles does not vary because of the heating. The quenching speed estimated at from simulations with the numerical code Fluent is greater than 40 000 K/s. So we check that the quenching is fast enough to avoid a combustion of carbon particles during dilution. The slight decrease in the number of particles after heating is likely to be a wall deposition by thermophoresis.

KCl alone (tests C and D)

Evaporation and re-condensation of KCl alone showed a difference between the cold tests and warm with dilution (Fig. 3). Indeed, when evaporation/re-condensation with dilution, the peak corresponding to the median diameter moved from 38.4 nm (without evaporation of KCl, so diameter of initial particles) at approximately 6.8 nm (indicia because it is close to the lower limit of detection of the SMPS) indicating the presence of a nucleation of new particles much finer. Thus, the evaporation / condensation tests of KCl alone highlight the fact that in the absence of particles in the flow, condensation of KCl goes through the formation of many fine nuclei.

KCl and carbon (tests B and E)

Fig. 4 shows the result of the condensation KCl in the presence of carbon particles. We can first notice the small amount of particles fine due to nucleation. It means that the almost all of the KCl condensed in the section test on pre-existing graphite particles or well in the form of deposits. Then, we can observe a variation of the diameter average of the collected particles. This diameter passes of 39 nm for graphite alone at 46 nm for the graphite on which condensed KCl. This corresponds well to the expected results: indeed, considering that the totality of the KCl was deposited on a population of spherical monodispersed particles of graphite of 39 nm of initial diameter, one calculates a final diameter of these particles of 52 nm .This is of course only a calculation of order of size, since the carbon particles are rather agglomerates than mere spheres.

Mass distribution of KCl during condensation

A series of three tests was conducted by measuring the amount of KCl deposited on the stages of the ELPI, each stage corresponding to a class of particle of fixed diameter. The aerosol generator used was the Palas AGF, which allows to obtain a diameter particles of KCl larger and therefore well distinct from that of graphite particles conditions of the three tests are gathered in the Table 2. Fig. 5 shows the results of the mass measurements, relative to the volume of gas circulating in the test section, collected on each stage of the ELPI. The first stage corresponds with the lowest aerodynamic diameters (<20 nm) and the twelfth largest large (> 5 μm). The measured mass distributions present several maxima: one for the initial KCl, two for the KCl condensed alone and three for condensed KCl in presence of carbon. These maxima correspond to modes of distribution that are the signature of different types of particles present.

The initial KCl (test no. 1) thus has a unimodal distribution centered on the stages 8 and 9 of the ELPI. This corresponds to an aerodynamic diameter of the order of 800 nm. This is the signature of non-evaporated KCl particles, here larger than those shown in Fig. 4 that were from a different generator. The biggest mode, centered on stages 8 and 9 for all distributions, corresponds to particles which have not been transformed during heating of the flow. This mode corresponds to 30% of the total injected KCl (ratio between the masses collected during tests 2 or 3 and those of test 1). In the case of condensed KCl without particles of carbon, a fine mode resulting from nucleation is well found on stages 2 and 3. This same mode is present for the KCl that condensed with party - carbon, and it is centered on the first stage corresponds to 10% of the KCl collected. This is the signature nucleated particles during cooling of the gas. Finally, the mode centered on stage 4 present only in the case of condensed KCl in the presence of carbon particles (test 3) is the signature carbon particles on which KCl condensed. It corresponds to about 50% of the mass collected KCl. We can notice that in total, if we add the different contributions for a total mass balance, about 50% of the injected KCl is collected on the trays of the ELPI. The remaining 50% remain in the test section in the form of deposit.

KCl deposits on the walls

Tests were carried out by measuring the concentrations of KCl present at the inlet and exit from the hot zone of the device. In Fig. 6, are represented the deposits at the walls of KCl as well measured on the device by input-output difference. These measurements show that the deposits on the walls decrease when the initial particle concentration carbon increases. We go from 40% in absence of carbon particles at 25% by injecting 3 × 1013 particles/m3.

Injection of particles and reduction of deposits

KCl deposits on the walls are due to two processes: the particle deposits on which KCl condensed, on the one hand, and condensation direct from the KCl vapor to the wall, other go. Deposits from the first process increase with the amount of carbon injected, while those from the second diminish. The trend decreasing observed in Fig. 6 results therefore of the predominance of the condensation process direct steam to the walls. In the absence of carbon particles in the flow, homogeneous condensation does not begin after nucleation and nucleation condensation. Before nucleation, so in the absence of solid nuclei, the temperatures of the walls are low enough to allow condensation direct condensable vapors to the wall. Thus, along the tube, before the appearance of nucleation, the vapors condense on the walls. By adding carbon particles in flow, as soon as the temperature of the gas is sufficiently weak, condensation on these particles becomes possible, at a higher temperature than that of homogeneous nucleation without particles of C. Thus, the area in which the KCl condenses only at the wall is greatly reduced. Thus, it seems possible to reduce the particles in an experimental facility in injecting particles into a flow containing condensable vapors before cooling.

VI. CONCLUSION

An experimental device has been designed to collect experimental data on the mechanisms condensation of aerosols in simplified conditions but representative of those encountered in a BtL process. Experimental results of condensation of KCl have highlighted the presence of nucleation when cooling the flow without any partials - carbon. This nucleation is always present by injecting carbon particles but then becomes negligible in the face of condensation by KCl growth on the initial particles. It has also been observed that the deposits on the walls condensable vapors are limited or even reduced by the presence of particles in the flow.

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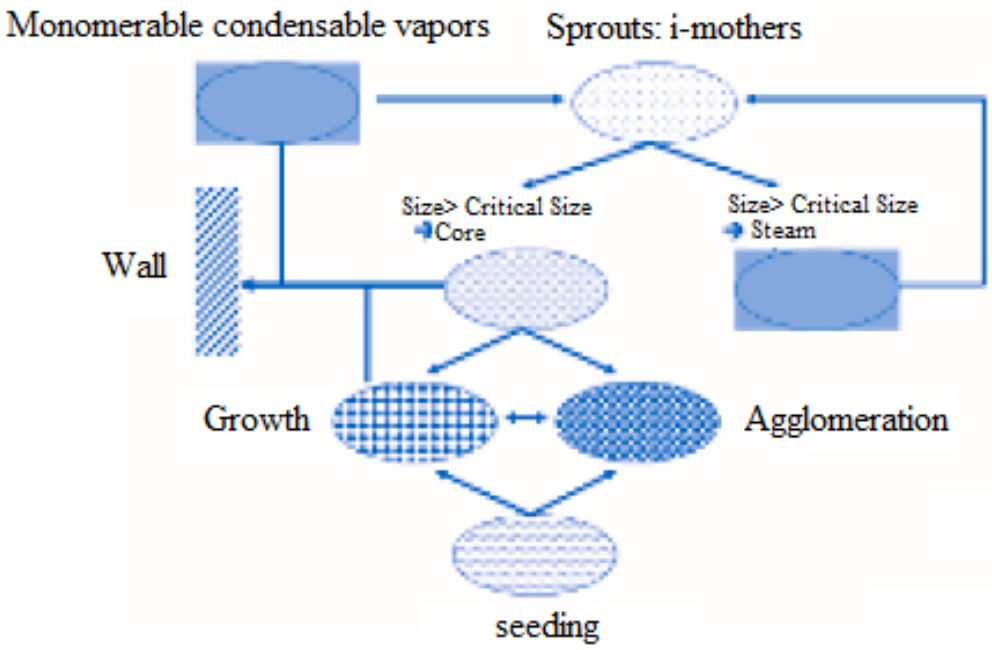

Fig. 1. General behavior of particles.

Table 1. Experimental conditions of preliminary tests.

Trial	Injected KCl concentration	Injected C concentration	Heating at 800 °C then cooling
A	0 mg/Nm^3	3×10^{13} /Nm^3	No (cold test)
B	0	3×10^{13}	Yes
C	4	0	No (cold test)
D	4	0	Yes
E	4	3×10^{13}	Yes

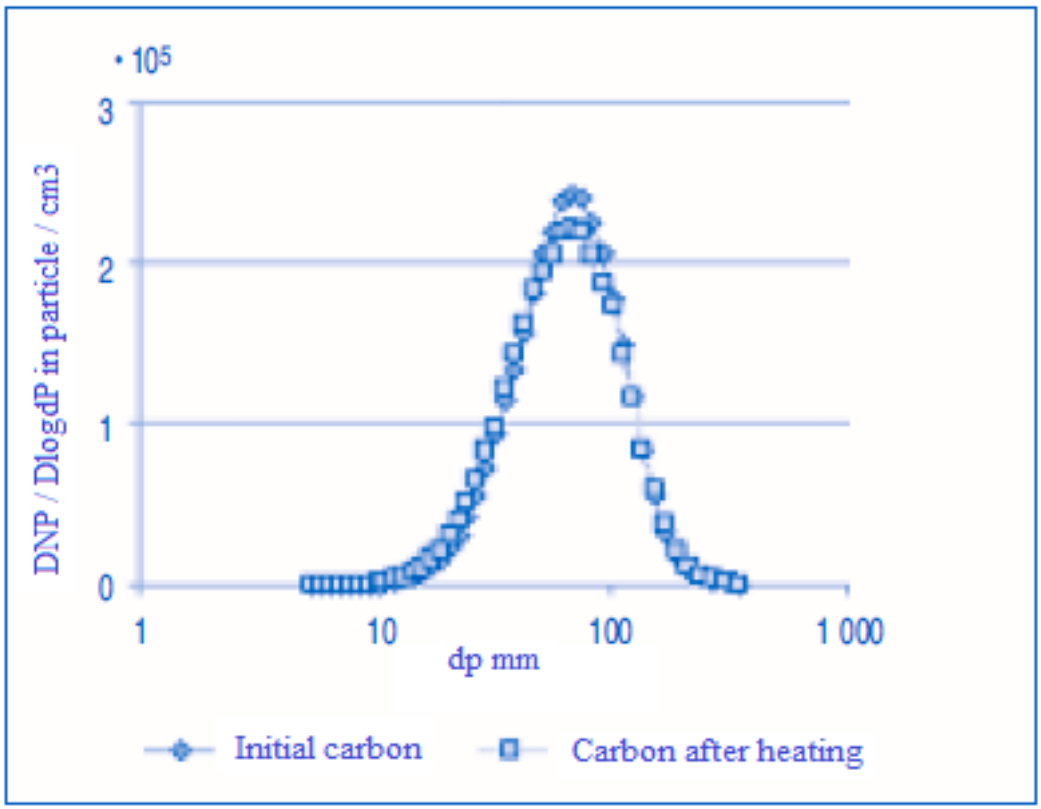

Fig. 2 Comparison of grain size distributions of carbon particles alone before heating and after heating (equivalent electric mobility diameter).

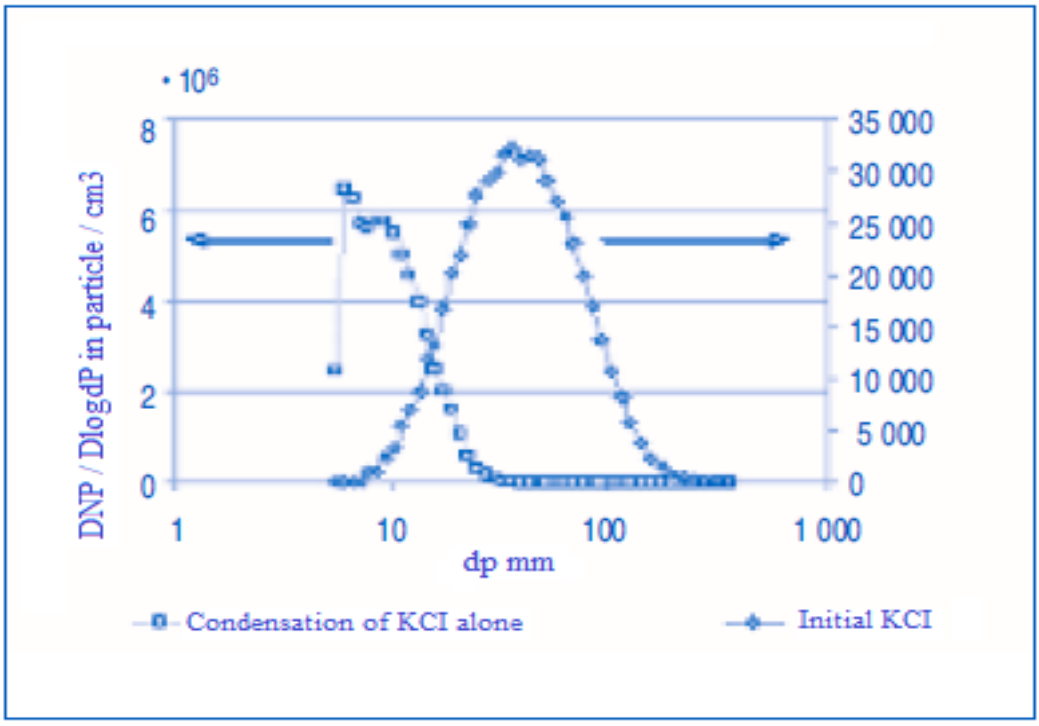

Fig. 3. Modification of the particle size distribution of KCl particles due to evaporation in furnaces (equivalent electric mobility diameter).

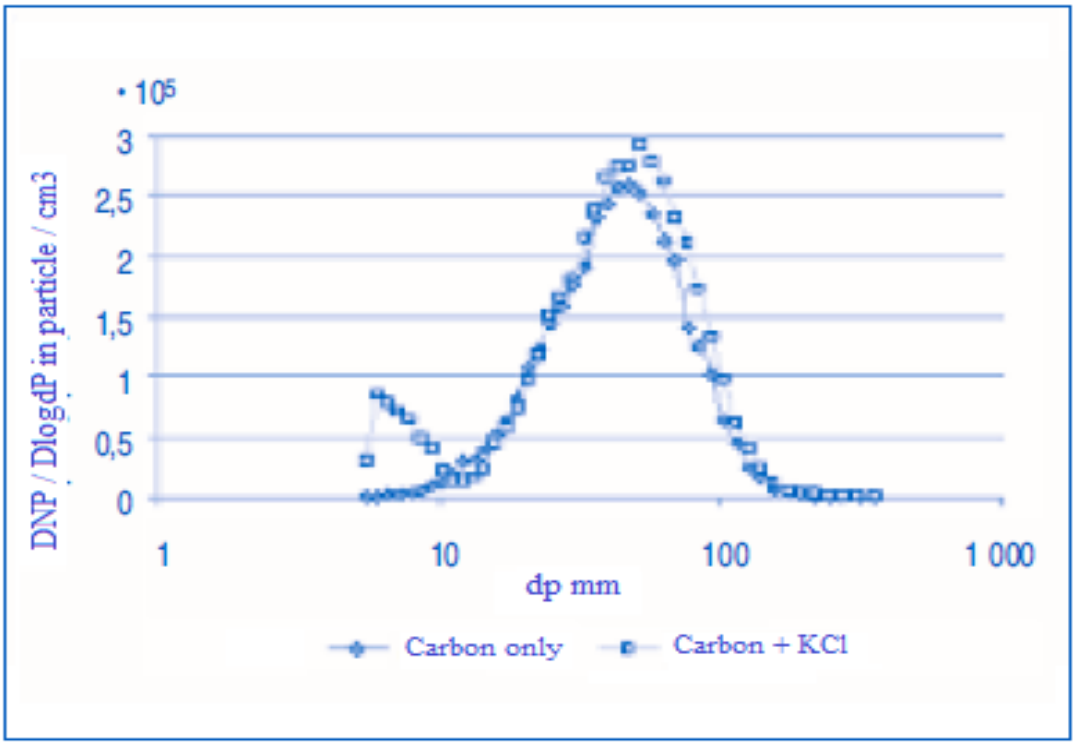

Fig. 4. Modification of particle size distribution due to condensation of KCl (equivalent electric mobility diameter).

Table 2.Experimental conditions of tests 1, 2 and 3.

Trial	Injected KCl concentration	Injected C concentration	Heating at 800 ° C then cooling
1	40 mg/Nm^3	0 /Nm^3	No (cold test)
2	40	3×10^{13}	Yes
3	40	0	Yes

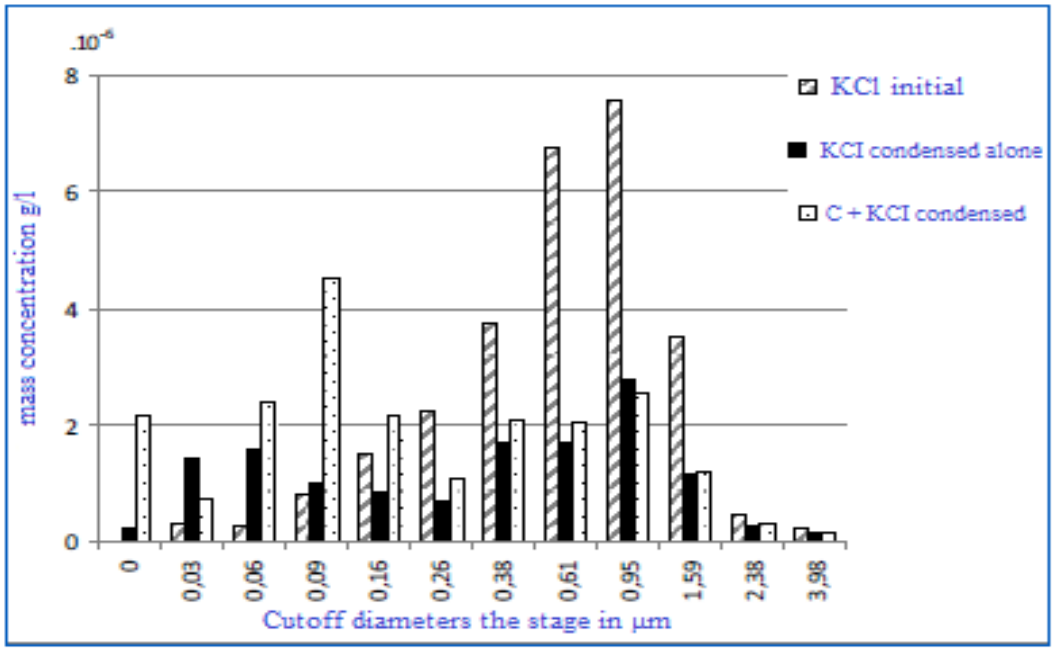

Fig. 5. Mass distribution of KCl as a function of the aerodynamic diameter of the particles.

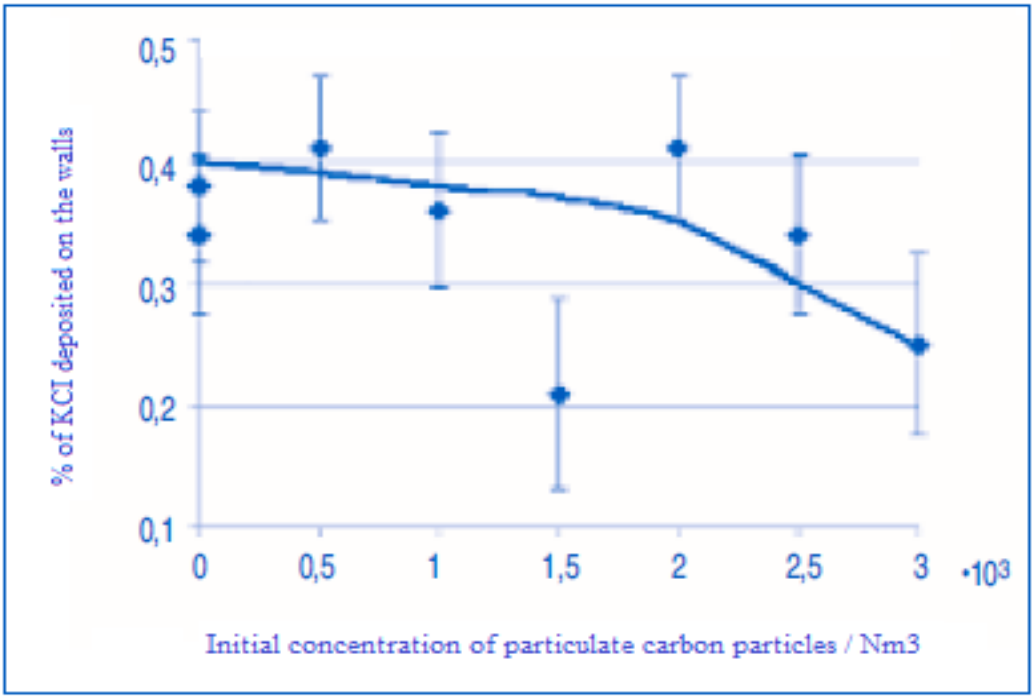

Fig. 6. Evolution of wall deposits as a function of the numerical concentration of initial carbon particles.