A NOVEL SEPARATION TECHNOLOGY FOR NANO PARTICLES AT DISCHARGE OF COMBUSTION AND INCINERATION EQUIPMENT

UNE NOUVELLE TECHNOLOGIE POUR LA SEPARATION DES NANO-PARTICULES A L’ECHAPPEMENT DES EQUIPEMENTS DE COMBUSTION ET INCINERATION

UNA NUOVA TECNOLOGIA DI SEPARAZIONE DELLE NANO PARTICELLE ALLO SCARICO DI IMPIANTI DI COMBUSTIONE E DI INCENERIMENTO

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Abstract

Still today, the issue of safely and efficiently avoiding the atmospheric release of the nano-particles produced by combustion and incineration processes is a critical and open challenge. This study addresses the conception, the technological realization and the first experimental testing of a new device suitable for in-duct filtration and separation of nano particles dispersed into flue-gas streams. The active filtering material is a membrane made from ptfe foil, in origin impermeable but suitable to allow creation, once properly stretched, of an inner texture of permeable micro- and nano-tubes, thus inducing activation of van der Waals effects to the advantage of improved particles’ sticking. The experimental tests confirm attainment of a remarkable filtration capacity, way better than the so-called ‘absolute filters’. Moreover, the filtration material allows to undergo a simple and safe “regeneration cleaning” process by which the particles can be re-collected off-duct without any filter dismantling.

Key-words: *nano particles in gas flows; membrane filter; nano particles separation; Ptfe membrane.*

Résumé

Le problème de la prévention des émissions à l'atmosphère des nano-particules produites par les procédés de combustion et de incinération est un défi toujours ouvert. L'étude présente la conception, la technologie de construction et les premiers essais expérimentaux d'un nouveau dispositif approprié pour la filtration et la séparation des nano particules dans les flux de gaz. Le substrat actif est une membrane en PTFE film: même si il est initialement imperméable, il permet la création, à l'intérieur, d'un réseau dense, mais perméable au gaz, de micro-et nano-tubes lorsque la membrane est soumis à une opération d'étirage planaire. De cette façon, sont également provoqué des actions induisant des effets de van der Waals à l'avantage de l’amélioration de l'adhésion moléculaire. Les tests expérimentaux confirment la réalisation d'une grande capacité de filtrage de micro- et de nano-

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particules, bien mieux que les ’filtres absolus’. En outre, le filtre à membrane peut être “régénéré” par des impulsions de pression, donc le particules sont remobilisés et séparés sans avoir à démonter le filtre.
Mots-clés: *nano particules dans les flux de gaz; filtre à membrane ; séparation des nano particules; membranes en ptfe*

Riassunto

Il problema di impedire la emissione atmosferica delle nano-particelle prodotte dai processi di combustione e di incenerimento rappresenta una sfida tuttora aperta. Lo studio riguarda la concezione, la realizzazione tecnologica e le prime prove sperimentali di un nuovo dispositivo atto alla filtrazione e separazione di nano particelle presenti in flussi gassosi. Il substrato attivo è una membrana realizzata in pellicola di PTFE: essa, in origine impermeabile, consente la creazione, al suo interno, di una fitta trama di micro- e nano-tubi se sottoposta ad un processo di stiramento planare. In tal modo vengono altresì indotti importanti effetti di adesione molecolare riferibili alle forze di van der Waals. Le prove sperimentali confermano il raggiungimento di una notevole capacità di filtraggio delle nano particelle, in modo nettamente migliore dei cosiddetti 'filtri' assoluti. Inoltre, la membrana filtrante consente di essere “rigenerata” in modo semplice e sicuro tramite impulsi di pressione, le particelle entro i micro-pori vengono ri-mobilizzate e separate senza smontare il filtro.
Parole chiave: *nanoparticelle in flussi gassosi; filtro a membrane; separazione delle nano particelle; membrane in ptfe*

Introduction

The issue of filtering the nano particles in order to counteract their ambient release, as produced by industrial combustion or incineration processes, is one of the most challenging tasks presently facing not only the manufacturers of air- and gas-cleaning equipment but also the scientific community active in this field. Indeed, many problems involved are still open, regarding theoretical, physico-chemical, technological and even diagnostic perspectives. A further aspect which some times adds up to this scenario is the need to separate the particles so to allow their continuous or periodical removal (without disassembling the filter), either for their disposal or for collecting them in view of performing most detailed off-line analytical characterizations. In these cases, the progressive embedding of the nano particles within the substantial thickness of porous filtering materials turns out as a major drawback when adopting thick-walled filters.
In gas filtration the basic principles of particle ‘mechanical’ deposition on filter media are known as screening, inertial impaction, direct interception and diffusion: the families of filters, typically made up of synthetic or glass fibers, which work by exploiting the said filtering actions, include panel filters, pleated filters, bag and pocket filters (Seeberger, 2011). Once the particles hit the fibres of the filter medium they are normally retained, however, often they can be released due to

unexpected shedding phenomena, a rather under estimated effect (Lehtimäki & Taipale, 2000) (Ginestet & Pugnet, 2008).

A different technology, by using appropriate synthetic materials, takes advantage of ‘natural’ electrostatic attraction forces between particle and fibre surface in order to strongly enhance the capture of nano particles. Electrostatic forces help to improve particle filtering efficiency without increasing the pressure drop of the filter media, thus meeting the requirement of a low energy demand during filter operation. However, although the effectiveness of these ‘electret’ filter media is good in many applications, the legitimacy of its use is still questioned as the electret effect may decay in a non-predictable way under real life conditions (Lehtimäki et al., 2005) (Hanley & Owen, 2005).

On the other hand, the high-voltage electrostatic precipitators are in general quite efficient in gas cleaning operations but less so in the nano range. In addition, the particles’ adherence to the tightly packed electrode plates makes it difficult to clean the plates as well as to separate and collect the nano particles. Ozone production by corona effect is another drawback of this technology (Ogawa, 1984).

In order to try overcoming the above limitations, a very recent ‘technologically engineered’ synthetic material was investigated in connection with its nano-particles’ filtering and separation capabilities, to be used directly within the discharge ducts of combustion and incineration equipment. The material, in shape of very thin membranes, is made from pure ptfe (polytetrafluorethilene), which gives the membranes the very interesting property of a permanent hydrofobicity: thus, they are not wetted by humid air, and the usual phenomenon of particles sticking and agglomeration on the surface or within the filter material is avoided. A peculiar feature of these very thin ptfe ‘foils’ is that they are natively impermeable, whilst they acquire a controllable permeability once properly engineered by subjecting them to a sort of planar stretching, so to induce formation of micro- and nano-tubes inside their inner structure (Sartorius, 2008). In the following, a first experimental application of these membranes as filtering medium for nano particles is discussed, in the case of discharge fumes produced by a small wood-pellet combustor, operated as a pilot incenerator.

A new filtering and separation technology: materials and methods

The INESE project (Impact of Nanoparticles in Environmental Sustainability and Ecotoxicity, web site: www.inese.it) is an on-going Italian interdisciplinary research programme sponsored by IIT (Istituto Italiano di Tecnologia), aimed at assessing the impact of engineered nanoparticles conveyed or sprayed into a test-greenhouse, wherein a suitable ecosystem is reproduced. To this end, a solid-fuel combustor device, actually a wood-pellet stove suitably re-arranged into a sort of pilot incinerator, has been installed, inside a small log cabin, in proximity of the greenhouse (sited near Modena), and its combustion fumes are directly discharged into it. In parallel, a second, ‘twin’, pilot combustor has also been installed at DIME/SCL (the Combustion Lab sited in Savona) where all the investigations here reported have been performed.

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Figure 1 - *The pilot combustor*

Figure 2 - *The pilot combustor (in the cabin) fumigating the greenhouse*

The functional characterization of the combustor, in terms of flame temperature as function of flue-gas flow rate is reported in Fig. 3. In order to limit the greenhouse heating, the lowest possible power-setting of the combustor was selected, corresponding to a flame temperature of 1045 °C and a flue-gas flow-rate of 63 m^3/h (Fig. 3).

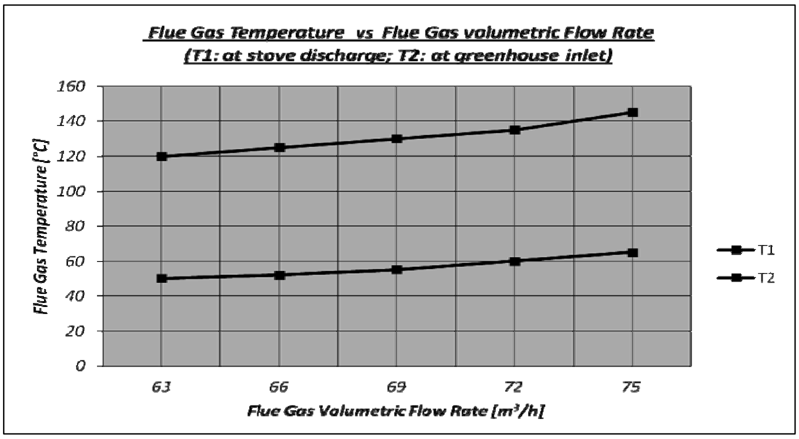

Figure 3

Flame temperature behavior as function of fumes' flowrates

The fumes' temperature, as attained after 9 m of tubing (distance between stove and greenhouse), was higher than 100 °C, unsustainable for the greenhouse environment. The provision of installing a water cooler along the fumes tubing appeared unfeasible, due to in-tubing massive humidity (and nanoparticles) condensing problems. Therefore, the solution of inserting an air blower for diluting, and mitigating, flue-gas temperature ('fumes forced-extraction') turned out as a quite efficient and effective provision. Figure 4 shows attainment of less than 50 °C at greenhouse fumes' inlet, a temperature at all manageable by the available air conditioning device.

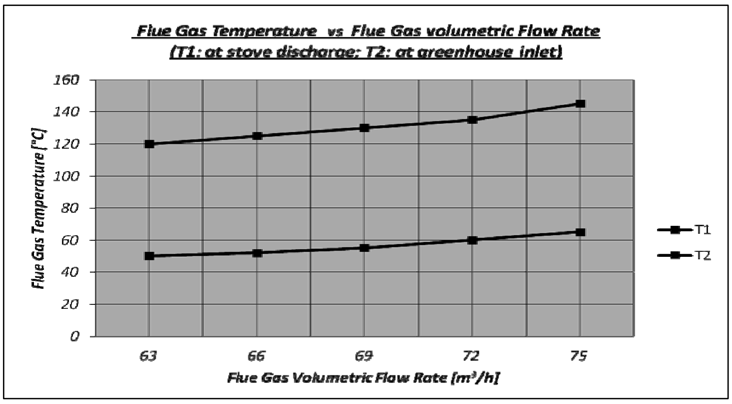

Figure 4

Fumes'temperatures at stove exit (T1) and at greenhouse inlet (T2)

Once completed the thermo-fluid-dynamical characterization of combustor's performance, greenhouse fumigation tests begun. In order to guarantee containment of nano-particles inside the tightly confined 'fumigation area' of the greenhouse, thus safely avoiding their ambient release, the greenhouse air-exchange ventilation windows were equipped with certified "absolute" filters, HEPA "Minilamp" type. By this means, any dangerous outside ambient escape of nano-particles should have been safely and strictly prevented, which actually was not achieved. Detailed measurements taken directly at the filters' downstream locations (outside of the greenhouse) pointed out a significant ambient release of nanoparticles, making mandatory the adoption of more effective filtering devices.
Owing to the above issues, the research was addressed at devising new provisions suitable to achieve the following critical targets: a) to avoid any uncontrolled ambient-diffusion of combusted nanoparticles by means of pursuing their in-duct filtering and separation; b) to 'selectively clean' the combustion fumes (from the micro-particles) so to achieve a kind of 'fumes-free' dispersion made up only of combusted particles of nano-dimensions; c) to safely manage the filtered/separated nanoparticles' collection for performing their off-line analysis, or else their safe disposal, or maybe even their subsequent controlled re-dispersion.
The issue of directly filtering, separating and collecting nano-sized particles within the discharge duct downstream of an incinerator is hard to tackle, due to the combined problems induced by:

- a very dense filter texture (in order to control nano-porosity)
- the in-duct higher velocities as compared with usual air ventilation openings
- the related high pressure drops
- the need to sustain temperatures above 100 °C
- a progressive blockage due to particles' build-up (very important!)

Since the above problems exclude any solution based on adoption of multiple-ply glass micro-fibered 'absolute' filters, at first, we resorted to using single-sheet (no-

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ply) membranes made of Cellulose Nitrate (Sartorius type 11407, White, 0.2 micron 'max' porosity, in figure 5) and also of Polyamide (Sartorius type 25007, White, 0.2 micron 'max' porosity). After several tests, we concluded that both these solutions had to be discarded mainly due to fouling and sticking of particles on the membranes' surfaces, inducing a rapid progressive increase of pressure drop. We attributed this behaviour to the hydrophilic nature of the membranes.

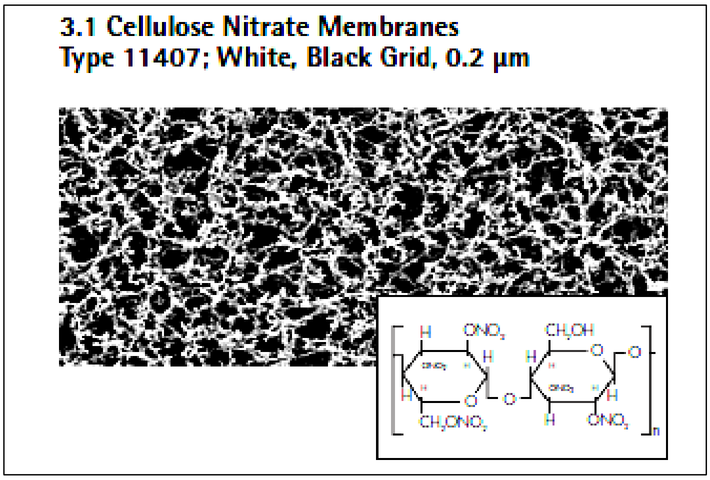

Figure 5

Structure of cellulose nitrate membranes

Owing to above reasons, a very recent 'engineered' material was selected, provided and tested: specifically, the Sartorius membrane type 11807 in ptfe (0.2 micron max porosity). Made from pure ptfe (polytetrafluorethilene), the membranes are permanently hydrofobic and thus are not wetted by humid air.

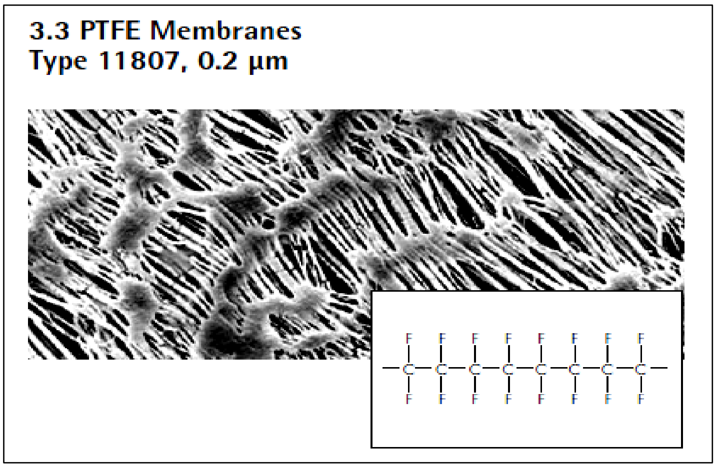

Figure 6

Structure of PTFE (polytetrafluor ethylene) membranes

A peculiar feature of these ptfe foils (0.065 mm thick) is that they are, in origin, impermeable, whilst a controllable permeability, made up of a uniform texture of inner nano-tubes, is induced into their structure once properly engineered by subjecting them to a sort of planar stretching. Quite interestingly, the membranes' hydrofobic behaviour, combined with the slippery aspect of their surfaces, turns out as strongly contrasting any particles' agglomeration or fouling build-up, to the advantage of a remarkable capacity of 'collecting' the nanoparticles within the nano-tubes of the membranes' inner structure. The filtering process takes unique advantage not only of the inner pores' nano dimensions but also of the combined effects of van der Waals intermolecular bonding actions, particularly effective for engineered (stretched) ptfe material once utilized in gas filtering applications at difference with liquids. A non secondary benefit which turns out from this quite interesting physico-chemical scenario is the capacity of the membranes to undergo 'regeneration cleaning' by subjecting them to 'up-flow' air pressure pulses, which opens the way to nano particles separation and re-collection with no need of dismantling the filters.

On the base of the peculiar characteristics of above membranes, an innovative device has been conceived and realized, aimed at efficient filtering, and possibly separation, of nanoparticles within discharge ducts of combustion and incineration equipment, allowing, if required, their safe collection for subsequent analysis. Due to the significant pressure losses induced by the filtering material, a multi-membrane (in parallel) configuration was adopted, with advanced-technology provisions for imparting high pressure resistance to the intrinsically weak membranes (in ptfe-sheet, 0.065 mm thick) if not properly supported: this has been achieved by means of high-precision laser multi-hole drilling of the inox-steel backing plates supporting each single membrane (each of 100 mm dia.).

To overcome the differential pressure drop a blower was provided ahead of the filter. The pictures in the following figures 7 through 12 show the manufacturing process of the multi-membrane filter, its mounting on a trestle and the tubing connections with the blower. The picture of the test-circuit is given in figure 10, showing the pellet-stove, the multi-membrane filter and the connecting piping. The blower is positioned behind the stove.

The fluid-dynamic performance tests of the circuit showed, in correspondence of the fumes' flow-rate suitable for greenhouse fumigation (63 kg/h), a pressure drop of about 190 mbar, which is significant, but consistent with the gas passages dimensions allowed by the nano-pore texture of the filtering material. The complete characteristic curve of the blower and the pressure drop behavior of the multi-membrane filter are given in figure 14: by adjusting the control valve on the circuit, a whole range of flue-gas flows (up to about 71 m^3/h) can be achieved, covering most of the combustor operating range.

Of course, particularly for industrial applications, in order to save energy by controlling pressure losses, higher filtering-area cross-sections will be required, which can be easily attained by using membrane-foils of suitable area dimensions.

Figure 7 – *Multi-hole drilling of single-membrane backing-plate*

Figure 8 – *Membrane holder with O-ring flanged backplate*

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Figure 9 – *Inner disk holding the multi-membrane arrangement*

Figure 10 – *Multi-membrane filter nearing completion*

Figure 11 – *Multi-membrane filter mounted on its trestle*

Figure 12 – *The filter connected to the blower*

Figure 13

Test circuit with pellet stove and multi-membrane filter. Blower is behind stove

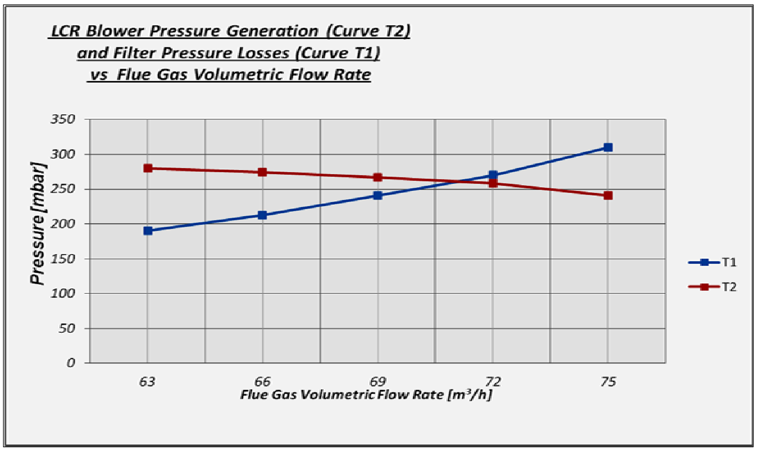

Figure 14

Filter's pressure losses vs blower's fluid-dynamic characteristic curve

Filtering performance analysis: diagnostic instrumentation, results and discussion

By taking advantage of the availability, at DIME/SCL in Savona, of a test-chamber aimed at burners' performance and exhaust gasses characterization (Fig. 15) , it has become conceivable to adapt this facility and to use its most advanced instrumentation for assessing, in real-time, the concentration, the number and the composition of nano particles as produced by the pellet stove here discussed when operated as a pilot incineration equipment.
To this end, its discharge tubing was directly connected to the inlet of the chamber by a sort of simple nozzle (a sort of fumes' thrower) slightly protruding into the chamber in place of the usual 'burner'.

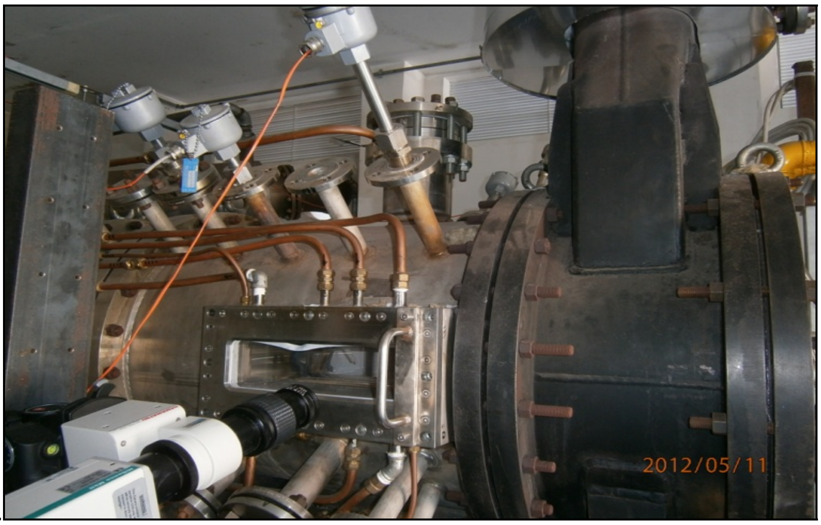

Figure 15

Test-chamber for flame and flue-gas analysis

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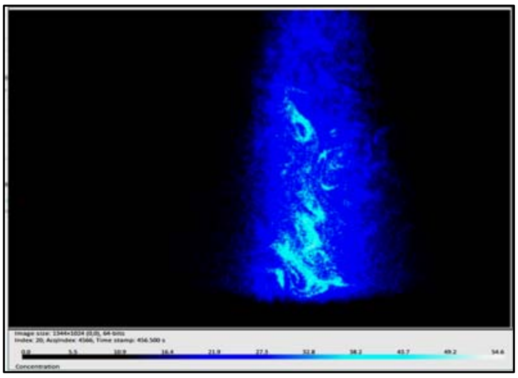
The instrumentation employed for performing the diagnostics of the particulate has been the available LIF system ('Laser Induced Fluorescence') equipped with an extremely recent add-on: an LII instrument ('Laser Induced Incandescence') (Figs. 16 and 17).

Figure 16 - *LIF image intensifier and CCD camera*

Figure 17-*LIF+LII images. CO vol. fraction: dark blue. Nano particles: light blue*



LII involves heating, directly in the combustion zone or in the discharge duct, any carbon-containing particles via laser radiation to the vaporization temperature, and measuring the resulting incandescence with a light-sensitive detector. More specifically, LII aims to measure the thermal emission (incandescent light) emitted from particles heated by a pulsed laser to temperatures in the 2500 K to 4500 K range. Quite interestingly, the resulting signals are sensitive to 'primary-particle' carbon mass concentration, independently of any subsequent particle aggregation.
The measurements have been performed, so far, with the pilot combustor fueled with standard pellet, without any addition of engineered nano particles.

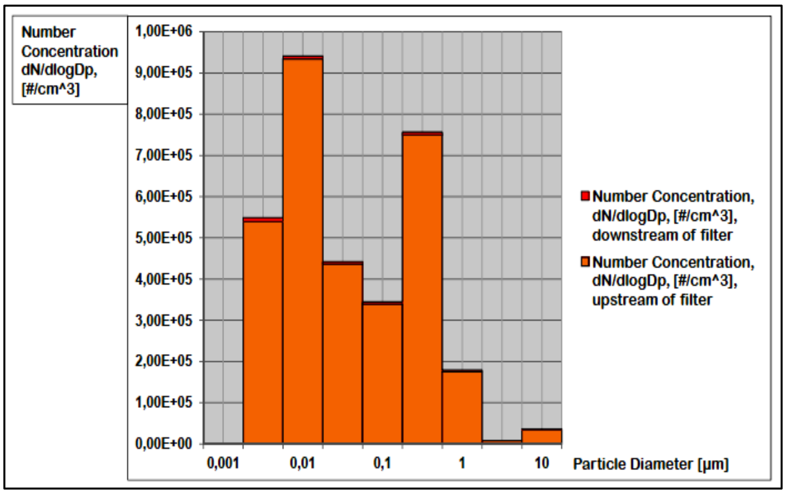


Figure 18

Particles' number concentrations as measured upstream and downstream of filter

The 'pellet+added particles' modality is already planned to be pursued, in a very near future, directly in the greenhouse test-site of Modena.
On the other hand, the evidence of nano particles' significant concentrations in the exhaust gasses of a pilot combustor fueled with plain pellet, without any added particles, was already ascertained during previous INESE test campaigns.
Therefore, it was decided to start testing the multi-membrane filter with the pilot combustor operating with plain pellet. The results, presented in figure 18, show the number concentrations of the nano particles as detected in two locations: the first, directly at the pellet-stove discharge (upstream of the filter); the second, downstream of the filter. Notice a remarkable capacity of the multi-membrane filter to abate, rather uniformly all along the wide range of particle diameters, the micro- and nano-particles produced by the combustor and released in the flue-gasses, from number concentration levels well into the hundreds of thousand and approaching one million for 10 nm particles, down to a few thousand at the most.
This outcome is expected to further improve for lower pressure drops, to be attained by increasing the membranes' surface area, and thus lowering the through-flow velocities at the filter. The capacity of the filter to abate particles with diameters well below its max porosity (set at 0.2 µm) is attributed to van der Waals effects.
In order to test the capacity of the filter to keep its performance for long operative times, a series of measurements were performed after different times of operation. In the case of 1 hour continuous operation, the corresponding results are shown in figure 19.

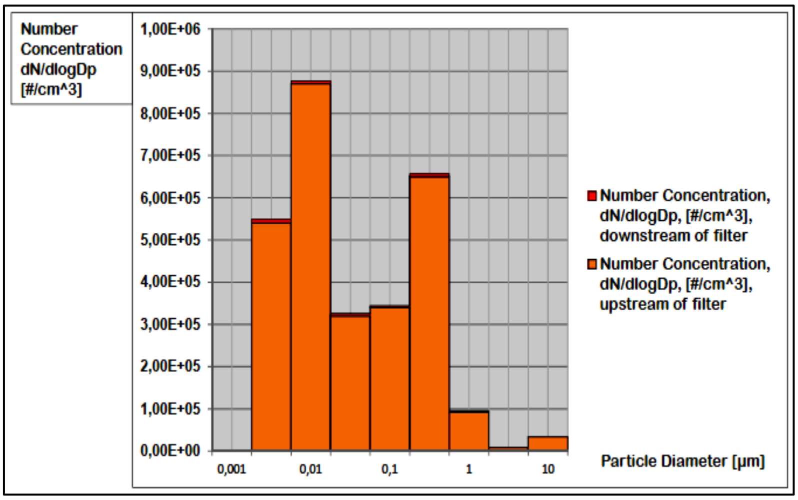


Figure 19

Particles' number concentrations as measured after 1 hour of filter operation.

Notice the excellent filter's capacity of keeping a non degrading performance. The pressure drop in correspondence of this condition increased by less than 5% with respect to initial situation. Although not yet investigated in detail, the possibility of periodically cleaning the membranes by applying counter-flow pressure pulses,

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thus 'regenerating' the filter's performance, has appeared at all feasible. By this means, the separation and collection of the 're-floating' nanoparticles, gathered, inside the filter, within the diverging duct located directly upstream of the membranes-holding disk, should become an easy and safe operation.

Conclusions

In this study, a new technology is presented and discussed suitable for in-duct filtration and separation of nano particles, as typically produced by industrial combustion processes and incineration equipment. The active filtering substrate is a highly hydrofobic ptfe foil ('membrane'), natively impermeable but suitable to allow creation of an inner texture of permeable micro- and nano-tubes once properly engineered by means of suitable stretching. The improved filtering capacity of micro- and nano-pores induced within a stretched ptfe material, as compared with other fiber material, is attributed to peculiar activation of van der Waals forces in presence of gaseous fluids. The experimental tests, although performed at a laboratory scale, appear at all encouraging because not only the filtration capacity turns out remarkably better than the so-called 'absolute filters' but, at difference with these latter, the filtration material allows to undergo a simple and safe 'regeneration cleaning' process by which the particles can be re-collected off-duct without any need of filter dismantling.

References

GINESTET A., PUGNET D. (2008) Shedding of particles from general ventilation filters: Assessment of test method developed by ISO/TC 142/WG3, CETIAT Technical Note, NTV 2008/088.

HANLEY J.T., OWEN M.K. (2005) Development of a New Conditioning Aerosol for Testing Electret Filters, ASHRAE Transactions, Symposia, 1190-RP, 1115.

LEHTIMÄKI M., SÄÄMÄNEN A., TAIPALE A. (2005) Investigation of Mechanisms and Operating Environments that Impact the Filtration Efficiency of Charged Air Filtration Media, 1189-TRP, VTT Technical Research Center of Finland.

LEHTIMÄKI M., TAIPALE A. (2000) Experimental study of particle shedding from ventilation filters, Ventilation 2000 Conference, Helsinki, Finland, 1:200-204.

OGAWA A. (1984) Separation of particles from air and gases, CRC Press, Boco Raton, OSTI 6134611.

SARTORIUS STEDIM BIOTECH (2008) Microfilters, Publ. SM-1503-e06124, Order No.: 85030-515-59.

SEEBERGER A. (2011) Synthetic filter media: Balancing energy efficiency and electrostatics in new synthetic filter media, Filtration & Separation, Elsevier 48(1):22–25.