The paper investigates the possibility to improve the filtering process of flue gas by controlling entrapment of suspended nanoparticle using dielectrophoresis (DEP). A realistic description of the manipulation process requires an accurate description of microchannel geometry and a precise evaluation of the DEP forces spatial distribution. The work presents the results of a numeric study which aims to characterize the functionality of a 3D DEP-based microsystem for the selective manipulation of nanometric particles. The analysis focuses on the nanoparticles having radii ranging from 50 to 150 nm, particles that cannot be filtrated by classical techniques but have a harmful effect for environment and human health. The solutions of the DEP force and particle concentration distribution for a typical separation device with interdigitated electrodes array are calculated using the COMSOL Multiphysics finite element solver. The performances of the device are analyzed in terms of a specific quantity related to the separation process, called Filtration efficiency. The simulations provide the optimal set of values for the control parameters of the separation process, and represent a useful tool in designing of microfluidic devices for separating nanoparticles from flue gas.

Key words: Air pollution, Flue gas filtration, Nanoparticle separation, Microfluidic device, Dielectrophoresis, Filtration rate, 3D numerical simulations.

1. INTRODUCTION

Filtration of submicron particles suspended in flue gas is an important technological challenge, as in urban environment the burning processes including incinerators of waste or diesel emissions are responsible for the emission of a significant amount of nanoparticles [1]. The studies on the impact of inhaled nanoparticles on the human health showed that the nanoparticles with size ranging from 50 nm to 200 nm are the most deleterious, due to their capacity to remain trapped in the inner respiratory ways or infiltrate into the blood. From a public health standpoint, the size of a particle is as important as its composition, recent research showing that although raw materials may not be dangerous, they can become toxic under the form of nanoparticles [2, 3]. Although the nanoparticles have smaller masses than microparticles, their number is at least four orders of
magnitude higher than the number of all other particles found in the flue gas. As a direct and immediate consequence, the filtration of nanoparticles is an important technological challenge, as they are produced in large quantities from material synthesis and combustion emission. Although the sources of polluting emissions are generally equipped with various filters, they are efficient only for the capture of the up to micron particles, but permit nanoparticles to escape in atmosphere [4, 5]. In the last decades, as technology is moving towards the nanoscale, several new methods of particle manipulation and filtration are being explored. At submicron scale, the classical mechanical devices of controlling particle movement proved to be ineffective, while the separation by flotation-based methods are usually slow and may contaminate the particles under manipulation. The so often used Corona electrostatic filters have a very high micrometric particle retention efficiencies (93–99%), yet most nanoparticles still remain undetected [5, 6]. In this context, the methods utilizing electric fields are emerging as most promising techniques for nanoparticle manipulation, in particular those based on dielectrophoresis (DEP). DEP is a phenomenon in which, under spatially non-uniform AC or DC electric fields, dielectric particles move because of the interaction of the dipole induced in the particle and the applied field gradient. DEP force does not require electrically charged particles; the strength of the force depends on the medium, particle’s electrical properties, particle’s shape and size, and on the applied electric field frequency [6, 7]. Since the relative dielectric polarization of the nanoparticles depends on the driving frequency of the applied electric field, an alternating (AC) electric field is usually applied to generate DEP forces of different magnitudes and directions. All the above mentioned reasons make the DEP-based methods well suited to be used for the control of position, orientation and velocity of submicron particles. In classical dielectrophoresis, the positive DEP force attracts particles into the regions of strong electric fields, while negative DEP force repels them from those regions [8, 9]. More recently, using advanced microelectrode fabrication techniques, the technology has moved into the submicron world so that nanoparticles can now be characterized and separated. The manipulation of nanoparticles in microsystems by using DEP forces has many existing and potential applications, presenting the advantages of voltage-based control and dominance over other forces: in the range above a few millimeters, the electrical forces are rather ineffective, but in the micron and submicron scale the electrical forces dominate [10].

This paper presents a study regarding the improvement of the filtering process by the entrapment of nanoparticles dispersed in flue gas in a microfluidic device using positive dielectrophoresis. The study exploits numerical simulations in order to investigate the behavior of nanoparticles with size ranging from 50 to 150 nm in a 3D DEP-based microsystem, which consists in a microchannel-working unit of a particulate trap. The numerical solutions of the positive DEP force and particle concentration distribution for a typical separation device with
interdigitated electrodes array are calculated using the COMSOL Multiphysics finite element solver. After a brief presentation of the theoretical model, we proceed to the experimental determination of the key characteristics of the particles suspended in flue gas: the size distribution of the submicron particles collected from the flue gas filters of a hazardous waste incinerator plant and their dielectric properties are evaluated. Based on both the experimental data and a proposed mathematical model, the concentration profile of nanoparticle suspension inside the microfluidic separation device is calculated and the performance of the device is analyzed in terms of a specific quantity correlated to the separation process, called Filtration efficiency. The final goal of the study is to identify a suitable way to predict accurately the particle entrapment by using numerical solutions of the DEP-flow equations for the fluid and find the most suitable values of the control parameters for separation process, in order to optimize the design of microfluidic devices for separating nanoparticles from combustion gases.

2. THEORETICAL CONSIDERATIONS

The time averaged DEP force acting on a spherical particle situated in an AC electric field can be describe by the following equation [11]:

$$\langle F_{DEP} \rangle = 2\pi a^3 \varepsilon_m K_R(\omega) \nabla \left( |\nabla V_R|^2 + |\nabla V_I|^2 \right),$$  \hspace{1cm} (1)

where we denoted by $a$ the particle radius, $\omega$ the angular field frequency, and $V_R$, respectively $V_I$ the real and imaginary part of the electric potential phasor, $\nabla = V_R + jV_I$, with $j = \sqrt{-1}$. For a homogeneous medium, the electric potential satisfy the Laplace equation $\nabla^2 \nabla = 0$. $K_R(\omega)$ is the real part of the complex quantity $\tilde{K}(\omega) = (\tilde{\varepsilon}_p - \tilde{\varepsilon}_m)/(\tilde{\varepsilon}_p + 2\tilde{\varepsilon}_m)$, named as Clausius–Mossotti (CM) factor, which is a measure of the effective polarizability of the particle. $\tilde{\varepsilon}_p$ and $\tilde{\varepsilon}_m$ are the complex dielectric permittivities of particle and medium. The complex permittivity is defined as $\tilde{\varepsilon} = \varepsilon - j(\sigma/\omega)$, where $\sigma$ is the electrical conductivity. Consequently, the CM factor can be expressed as [12]:

$$\tilde{K}(\omega) = \frac{(\varepsilon_p - \varepsilon_m) + j/\omega \cdot (\sigma_p - \sigma_m)}{(\varepsilon_p + 2\varepsilon_m) + j/\omega \cdot (2\sigma_m) \setminus},$$ \hspace{1cm} (2)

The CM factor depends on the dielectric properties of the particle and medium and on the frequency of the applied field; at low frequencies the sign is determined by the electrical conductivities of the particle and the medium and at higher frequencies by the permittivities. The variation in this factor results in a
frequency-dependent dielectrophoretic force that is unique for a particular type of particle (Figure 1).

![Graph showing variation of K_(R) with frequency](image)

Fig. 1 – Variation with frequency of the real part of the Clausius-Mossotti factor, \( K_R(\omega) \).

It follows naturally from these considerations that the DEP force can be used as an effective tool for separating particles, based solely on their dielectric properties and size. When the sign of the real part of the CM factor, \( K_R \), is positive, the particles are attracted to the locations of electric field intensity maxima and repelled from the minima, phenomenon known as positive dielectrophoresis (pDEP). The opposite occurs when \( K_R \) is negative, situation referred to as negative dielectrophoresis (nDEP).

Figure 2 illustrates a typical DEP-based separation device with parallel interdigitated bar electrodes placed on the bottom surface used in dielectrophoretic selective manipulation of submicron particles [7].

![Diagram of DEP patterning chamber](image)

Fig. 2 – Sketch of the DEP patterning chamber with interdigitated bar electrodes at bottom surface of the experimental device used for dielectrophoretic separation.

Due to the symmetry of the geometry and considering the electrodes much longer than their width, very often the problem is treated in two dimensions and the
electrodes' height is neglected. As a consequence of the periodic distribution of the electrodes, the numerical calculations of the DEP force and the concentration field can be performed considering as computational domain only a so called “basic unit cell”, which fully describes the entire system, except the vicinity of the walls. The geometry of the computational domain and the associated boundary conditions necessary to solve the Laplace equation for the electric potentials, \( V_R \) and \( V_I \), are presented in Figure 3.

Fig. 3 – Schematic representation of the computational domain with the associated boundary conditions for the real part (\( V_R \)) and imaginary part (\( V_I \)) of the electric potential. The solid lines indicate the basic unit cell [7].

Moving now to a upper length scale, the macroscopic behavior of a suspension of small spherical particles of radius \( a \) in a fluid of viscosity \( \eta \) is governed by the following system of equations [7]:

\[
\mathbf{v} = \mathbf{u} + \frac{2a^2}{9\eta} \mathbf{F}, \tag{3a}
\]

\[
\frac{\partial C}{\partial t} + \nabla \cdot \mathbf{j} = 0, \quad \text{where} \quad \mathbf{j} = C \mathbf{v} - D \nabla C. \tag{3b}
\]

where \( \mathbf{u} \) and \( \mathbf{v} \) are the fluid and particle velocities, \( \mathbf{F} \) is the external force acting on the particles (DEP force in this case), \( t \) is the time, \( \mathbf{j} \) is the particle flux, \( D \) is the diffusion coefficient of the particles, and \( C \) is the particle volume concentration.

The fluid flow field inside the separation device, \( \mathbf{u} \), is calculated by solving the classical Navier-Stokes equation in the compressible case, together with the corresponding boundary conditions. For the obtained DEP-force and fluid flow
field, the particle concentration is evaluated by numerically integrating equations (3a) and (3b). The calculated particle concentration field gives information at a local scale, showing how the particles are attracted on the margins of electrodes and the influence of the main parameters of the problem on this process. For the evaluation of efficiency of the filtration process we define a quantity named \textit{Filtration efficiency} (FE), which describes globally the process in terms of nanoparticles entrapment at the electrodes, as a function of the concentration distribution:

$$FE = \frac{C_{\text{input}} - C_{\text{output}}}{C_{\text{input}}} = 1 - \frac{C_{\text{output}}}{C_{\text{input}}} \, [\%]$$

(4)

where $C_{\text{input}}$ and $C_{\text{output}}$ are the mean concentrations of suspended nanoparticles at the input and the output surfaces of the device, respectively, as schematically represented in Figure 4.

![Figure 4](image)

Fig. 4 – The elements of the separation device used for defining the \textit{Filtration efficiency}.

This proposed quantity gives the global information on the filtration process, and can be used in order to evaluate the efficiency of the filtration process.

At the end of this theoretical section is important to conclude that the electrokinetic forces depend in a complex manner on system dimensions, frequency, field, etc. In separation systems, the buoyancy force can be sometimes significant (as in FFF) but often the magnitude of this force is much lower than the other forces acting on nanoparticles.

3. RESULTS AND DISCUSSION

3.1. EXPERIMENTAL RESULTS

In order to obtain relevant input data for the simulations, we investigated three probes, named as A, B and C, consisting in samples of ash resulted from the
combustion of different wastes, collected monthly from filters of Pro Air Clean Timisoara hazardous waste incinerator, within a period of three months. These probes were analyzed from dimensional and dielectric point of view.

For dimensional characterization, we prepared for each of the three probes a mixture of 5 mg ash in 100 ml distilled water at room temperature, and put it to rest for 20 minutes, in order to decant the microparticles. Then we collected the remained slurry liquid and analyzed the particle size/concentration distribution using a Nano Sight LM 10 nanoparticle visualization system, based on nanoparticle-tracking analysis method. Figures 5a-c illustrate the particle size/concentration distribution for the three probes. The distribution diagram indicates that it contains four significant groups of nanoparticles, having sizes of 55 nm, 100 nm, 155 nm, and 275 nm, respectively.

These diagrams show that the gas resulting from the combustion of waste contains nanoparticles, nanoparticles having radii ranging from 50 to 200 nm, potentially harmful for human health. Recall that these nanoparticles are only those that probably were attached to larger particles, and stocked during the mechanical filtering process. For each of the three probes one observe higher concentrations for particles with radii of about 50, 100, and 150 nm, values that will be considered relevant in our future analysis.
The frequency dependence of the real part of the CM factor for the mentioned powder sample was determined by dielectric measurements and using the Maxwell-Wagner model [12]. The frequency dependence of dielectric permittivity of the samples was determined in the frequency range 25Hz–2MHz, using a RLC-meter Agilent type E4980A, to which a cylindrical capacitor containing the analyzed material was connected. Figure 6 shows the calculated frequency dependence for the real component of the real part of CM factor of the probes A, B and C [12].

One observes that in the investigated frequency range, the behavior of the real part of the CM factor is described by three distinct variation regions: first, it decreases slightly for frequencies less than 200 Hz, then falls sharply for frequencies between 200 Hz and 4 kHz, and finally remains nearly constant for higher frequencies. Due to the positive values of the real part of CM factor, it follows that the use of positive DEP force is appropriate for manipulating the ash nanoparticles from the flue gas. For further analysis regarding the filtration process, only devices working at frequencies up to 200 Hz are of interest, because in this domain the CM factor has the highest value, \( K_R = 1 \), and consequently one reaches the highest strength of pDEP force acting on nanoparticles in the flue gas. In further calculations we use the value \( K_R = 1 \) for the real part of the CM factor.

3.2. NUMERICAL RESULTS

The experimental results obtained by dimensional analysis and dielectric measurements are used as input data for the simulation of the transport phenomena.
inside a realistic DEP device for nanoparticle trapping from flue gas. First we compute the pDEP force distribution inside a typical DEP device and then we move to the problem of determining the distribution profile of nanoparticles under the influence of dielectrophoresis. Finally, we analyze and discuss the obtained numerical results in terms of Filtration rate, a global quantity correlated with the concentration field, which offers a more suggestive characterization of the capabilities of the device regarding the separation process of nanoparticles from flue gas. All the numerical simulations were performed using the COMSOL Multiphysics program.

For the computation of the pDEP force, we first solved the Laplace equation for the real and imaginary components of the electric potential, together with the associated boundary conditions presented in Figure 3. The computational domain consists of a unit cell described by the following set of geometric parameters: $d = l = 100 \mu m$, $H = 500 \mu m$ and $w = 2 \mu m$. The simulations were performed for a suspension of particles with characteristic sizes $a = 50$ nm, $a = 100$ nm and $a = 200$ nm respectively, in air. The dielectric response of the particles is characterized by the real part of the CM factor $\Re K = 1$ and we considered the amplitude of the electric potential applied on the electrodes in the range $V_0 = 12 \pm 24 V$.

In order to avoid numerical difficulties, due mainly to the extremely wide range of variation of the DEP force inside the computational domain, we chose to solve the model equations in the dimensionless form. If the electric potential is scaled with the applied electrode voltage $V_0$, the distances with the electrode width $d$, the time with $d^2 / D$, the velocities with $D / d$, and the particle volume fraction with the initial average volume fraction $C_0$, the corresponding dimensionless form of the DEP force (1) is:

$$\langle F_{DEP} \rangle = F_{0DEP} \nabla' \left( | \nabla' V' |^2 + | \nabla' V'' |^2 \right).$$

We noted in the above equation $F_{0DEP} = 2\pi a^3 \epsilon K (V_0^2 / d^3)$ a quantity that measures the intensity of the external field. The prime symbol above denotes the dimensionless quantities.

The magnitude of the vector $\nabla' \left( | \nabla' V' |^2 + | \nabla' V'' |^2 \right)$, proportional to the dimensionless DEP force given by (5), calculated in the vicinity of the electrodes, is presented in Figure 7a in logarithmic scale. The results clearly show that the pDEP force reaches its maximum near the electrodes' margins (a difference of at least two orders of magnitude), and diminishes rapidly with the height. Practically, at heights $y > d$ the dielectrophoretic effect is negligible. The nanoparticles subjected to the calculated pDEP force are strongly attracted to the electrodes edges, while their volume concentration in fluid diminishes, as showed in Figure 7b.
The efficiency of the filtration process can be evaluated by calculating the Filtration efficiency given by relation (4) for different values of problem's parameters. The computation is performed using an iterative procedure: the output concentration in one unit cell is considered the input concentration for the next unit cell, in order to describe the cumulative effect of the filtration inside the dielectrophoretic device. This type of analysis allows an estimation of the necessary number of cells (or electrodes) in order to obtain a certain desired filtration rate, when the other parameters of the problem are fixed. The results presented in Figure 8a show that in the case of particle having size of 100 nm, a desired filtration rate of 90% can be obtain by using about 30 electrodes when applying a voltage of 24 V, about 60 electrodes for 18 V, and nearly 200 electrodes for an applied voltage of 12 V.

Fig. 7 – a) The magnitude of the vector $V'\left(\left|V'V'_n\right|^2 + \left|V'V'_l\right|^2\right)$, in logarithmic scale and b) Calculated particle concentration distribution for a typical separation device.
Filtration of flue gas in microfluidic devices using dielectrophoresis

Fig 8 – Calculated Filtration efficiency versus number of cells for a) particles with $d = 100$ nm at three different applied voltages and b) particles with three different radii at a fixed applied voltage of $V_c = 18$ V ($d = l = 100 \mu m, w = 2 \mu m$).

When we analyze the effect of particle radii on the filtration capacity, the results presented in Figure 8b predict that, for example, when the applied voltage is 18 V, particles of 150 nm are completely captured after 10 electrodes, for particles of 100 nm we need about 150 electrodes for the complete capture, while for the particles of 50 nm are captured less than 60% even if one use devices with 250 electrodes.

In conclusion, the simulations performed in the frame of the presented mathematical model allow an estimation of the performances of the filtration as a function of the geometric and physical parameters of the problem.

The next step of our research activity will focus on the validation of the proposed model. Some preliminary but promising experimental results were
already obtained. The tests performed with a DEP-based separation device having $l = d = 100\, \mu m$ and $H = 1000\, \mu m$ reveal that in the absence of the applied voltage the particles are not at all attracted to the electrodes (Figure 9a), while once applied an AC voltage of 12 V the dielectrophoretic effect appears. More than that, the concentration of captured particles clearly diminishes while we depart from the input region, which is in concordance with our simulations.

Fig. 9 – DEP-based separation device with $l = d = 100\mu m$ a) before fumigation, b) after fumigation, successive snapshots, $U = 12\, V$, AC, 50 Hz, time of fumigation $t = 30$ s.

After this qualitative validation, a quantitative evaluation of the concentration of nanoparticles captured at the electrodes, at different distances from the input of the device, is necessary in order to give a solid validation of the model.
4. CONCLUSIONS

This paper presents a 3D numerical study of a DEP-based microsystem for the selective manipulation of submicron particles using dielectrophoresis. The DEP force depends on the gradient of the energy density, which changes on the length scale of the electrodes and is a short range effect. It can be modulated by changing the frequency and electrical properties of the suspending medium. We introduced a theoretical model and applied it to a physically realistic problem, and then we predicted a novel mechanism of particle entrapment, which is not only fundamentally different from the one conventionally known but also, is proven to be efficient at small scales. The numerical solutions of the DEP force and particle concentration distribution for a typical interdigitated electrodes array are calculated using the proposed theoretical model. The efficiency of the manipulation process is discussed in terms of filtration efficiency, for different values of particle radius and applied voltage. The optimal parameters of the separation process can be determined through a detailed numerical study performed in the frame of the proposed mathematical model and used for designing more efficient separation devices.

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REFERENCES