THE CLAUSIUS-MOSSOTTI FACTOR IN LOW FREQUENCY FIELD OF THE POWDERS RESULTED FROM WASTES COMBUSTION

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Using the complex dielectric permittivity measurements in the frequency range 25 Hz to 2 MHz, the frequency (ω) dependence of the Clausius-Mossotti factor, was determined $K(\omega)$ for some ash powder micro/nanoparticles samples, which resulted from the combustion processes of municipal wastes.
The results show that if the ash powder is dispersed in air, the real part of the Clausius-Mossotti factor $\mathrm{Re}[\tilde{K}(\omega)]$, for all samples and at all frequencies in the investigated range is positive. Therefore, filtering of flue gas using positive dielectrophoresis (pDEP) is possible by trapping the nanoparticle in the area of the strongest electric field.
The theoretical study which was performed in this paper shows that the $\mathrm{Re}[\tilde{K}(\omega)]$ may change from a positive to a negative value, at a critical frequency, f_c (which decreases from 1.416 kHz to 0.31 kHz), if the dielectric permittivity of the dispersion medium increases from 3 to 10.
Therefore, there is a shift from positive dielectrophoresis (pDEP) to negative dielectrophoresis (nDEP), and the particles can move from regions of high electric field gradient to regions of low electric field gradient.
The results obtained show that there is the possibility of using the dielectrophoresis, for retaining through manipulation and spatial separation controlled by the nanoparticles from the powder samples investigated, leading to purification of exhausted combustion gases and reduction of the air pollution.

Key words: Complex dielectric permittivity, Clausius-Mossotti factor, Nanopowders, Dielectrophoresis, Air pollution.

1. INTRODUCTION

Dielectrophoresis (DEP) is one of the technologies which exploits the differences in the dielectric properties of particles, to allow manipulation and their characterization in a fluidic medium [1]. A non-uniform electric field, induces the movement of dielectrically polarized particles. The amplitude and direction of the dielectrophoretic force are dependent on the electric field, on the size of the particles and the electrical properties of the particles relative to those of the surrounding medium [2]. At a microscopic scale, the dielectrophoretic force, acting on a spherical particle of radius a, of complex dielectric permittivity $\tilde{\varepsilon}_p$, suspended in a medium of relative permittivity ε_m, is given by [3, 4]:

$$\vec{F}_{DEP} = 2\pi\varepsilon_0\varepsilon_m a^3 \operatorname{Re}\left[\tilde{K}(\omega)\right]\nabla E^2 \tag{1}$$

In this equation, ∇E^2 is the gradient of the square of the electric field, E being the magnitude (RMS - root mean square) of the applied field; ε_0 is the permittivity of free space and $\operatorname{Re}\left[\tilde{K}(\omega)\right]$, is the real component of the complex Clausius–Mossotti (CM) factor [5], given by:

$$\tilde{K}(\omega) = \frac{\tilde{\varepsilon}_p - \tilde{\varepsilon}_m}{\tilde{\varepsilon}_p + 2\tilde{\varepsilon}_m} \tag{2}$$

In Eq. (2), $\tilde{\varepsilon} = \varepsilon' - i\varepsilon''$, is the complex dielectric permittivity (subscripts p and m denote the particle and the suspending medium, respectively); $\omega = 2\pi f$ is the angular frequency of the applied field (f – frequency of the field); $i = \sqrt{-1}$ and ε' and ε'', are the real and imaginary components of the complex dielectric permittivity, which may be written in the Debye form [6]:

$$\varepsilon' = \varepsilon_\infty + \frac{\varepsilon(0) - \varepsilon_\infty}{1 + \omega^2\tau^2} \tag{3}$$

$$\varepsilon'' = \frac{\sigma}{\omega\varepsilon_0} + \frac{\left[\varepsilon(0) - \varepsilon_\infty\right]\omega\tau}{1 + \omega^2\tau^2} \tag{4}$$

where σ is the electric conductivity of the particle or medium and τ is the relaxation time. From the Debye theory [6] it is known that the relaxation time is correlated to the frequency f_{max}, at which ε'' has a maximum, by the relation, $2\pi f_{\max}\tau = 1$. Also, in Eqs. (3) and (4), $\varepsilon(0)$ is the permittivity at frequencies much smaller than f_{max} and ε_∞ is the permittivity at frequencies much larger than f_{max} [7]. For systems with more than one relaxation time, both equation (3) and equation (4) are sums of terms corresponding to different relaxation processes [7].

By introducing in Eq. (2) the complex form of dielectric permittivity of particle, $\tilde{\varepsilon}_p = \varepsilon'_p - i\varepsilon''_p$, respectively, the dielectric permittivity of medium, $\tilde{\varepsilon}_m = \varepsilon'_m - i\varepsilon''_m$, we can determine the real, $\operatorname{Re}\left[\tilde{K}(\omega)\right]$ and imaginary, $\operatorname{Im}\left[\tilde{K}(\omega)\right]$ components, of the CM factor. Result following relationships:

$$\mathrm{Re}\left[\tilde{K}(\omega)\right] = \frac{\varepsilon'^2_p + \varepsilon''^2_p + \varepsilon'_p\varepsilon'_m - 2\varepsilon'^2_m + \varepsilon''_m\left(\varepsilon''_p - 2\varepsilon''_m\right)}{\varepsilon'^2_p + \varepsilon''^2_p + 4\varepsilon'_p\varepsilon'_m + 4\varepsilon'^2_m + 4\varepsilon''_m\left(\varepsilon''_p + \varepsilon''_m\right)} \quad (5)$$

$$\mathrm{Im}\left[\tilde{K}(\omega)\right] = \frac{3\varepsilon''_p\varepsilon'_m - 3\varepsilon'_p\varepsilon''_m}{\varepsilon'^2_p + \varepsilon''^2_p + 4\varepsilon'_p\varepsilon'_m + 4\varepsilon'^2_m + 4\varepsilon''_m\left(\varepsilon''_p + \varepsilon''_m\right)} \quad (6)$$

From equations (5) and (6) we can observe that the real and imaginary components of the Clausius-Mossotti factor, depend on the dielectric properties of the particle and medium, and on the frequency of the applied field. The frequency dependence of this factor determines the dependence in frequency of the DEP force, which is unique to a particular particle type. Therefore, we can use dielectrophoresis as an effective means for particle separation, solely according to their dielectric properties and size. Being a measure of the relative permittivity between the particle and the surrounding medium, the CM factor determines the sign of the DEP force: when $\mathrm{Re}[\tilde{K}(\omega)] > 0$, the particle is more polarizable than its surrounding medium and is attracted toward the locations of maximum electric field intensity and repelled from the zones of minimum, phenomenon known as positive dielectrophoresis (pDEP). The opposite occurs with $\mathrm{Re}[\tilde{K}(\omega)] < 0$, referred to as negative dielectrophoresis (nDEP).

Using the positive/negative DEP feature and the size dependence of DEP force, dielectrophoresis (DEP) has been applied extensively in the separation and isolation of particles/cells [1, 2].

In this paper, we present a preliminary study regarding the possibility to capture the nanoparticles exhausted by combustion gases using dielectrophoresis, in order to improve the filtering processes.

In our study we considered that, for a particle exposed to AC electrical fields of different frequencies, the analysis of the DEP force reduces the frequency analysis dependence of the Clausius–Mossotti (CM) factor. Therefore, by measuring the frequency dependence of the complex dielectric permittivity, in the frequency range 25 Hz–2MHz, we determined the real part of the CM factor for a nanopowder sample obtained from combustion of wastes, taken from the flue gas filters of the mentioned incineration plant.

2. SAMPLES AND EXPERIMENTAL INVESTIGATIONS

The sample used for dielectric measurements was an ash powder containing a mixture of micro/nanoparticles resulted from combustion processes of the wastes, in the incinerator plant for municipal and hazardous wastes of the Pro Air Clean Timisoara. The incineration procedure is based on the complete burning of waste at 850–1000°C and post-combustion at temperatures between 950–1300°C of the resulting gas, followed by sudden cooling, a process that ensures the complete destruction of dioxins, furans and other toxic components resulting from special waste incineration. The plant allows any type of solid waste incineration (excluding radioactive wastes) in the inferior chamber, and liquid waste incineration such as pesticides, solvents, oils etc., through direct injection in the post-combustion chamber. Although the technological process is automatic and managed for the advanced control of emissions in the atmosphere, it cannot filter and control the nanoparticles contained in flue gas [8].

For dimensional characterization of the sample, we prepared a mixture of 5 mg ash powder and 100 ml distilled water at room temperature. After the decantation of micrometer particles, we collected the remained slurry liquid and then the particle size/concentration distribution was analyzed by using a Nano Sight LM 10 nanoparticle visualization system. This device determines the size distribution of nanoparticles in polydisperse and heterogeneous systems, using the method nanoparticle-tracking analysis (NTA). Figure 1 presents the analysis results regarding the particle size/concentration distribution for investigated mixture sample.

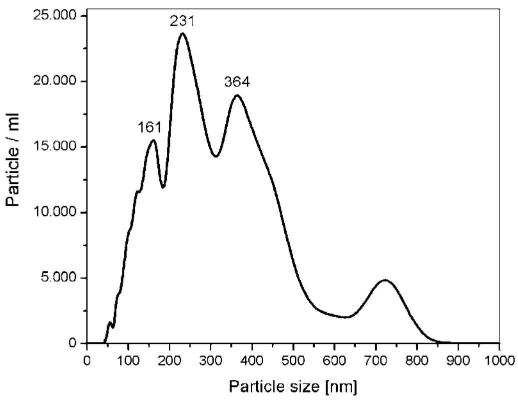
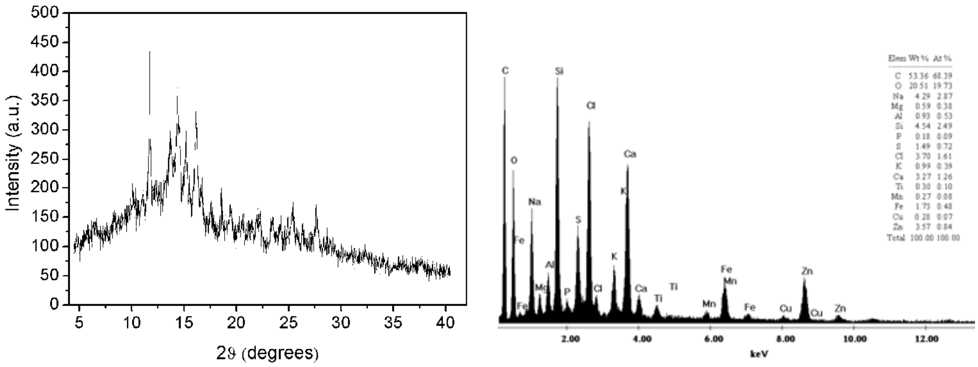


Fig. 1 – The particle size/concentration diagram.

As seen in figure 1, we obtained three significant particle sizes groups in the sample, at 161 nm, 231 nm and respectively 364 nm values.

The phase composition of the ash powder was determined by X-ray diffraction (XRD) using a Bruker D8 Advance System (Mo–Ka radiation, Zr filter) operating at 50 kV and 40 mA. Figure 2 a) shows the XRD spectrum for the investigated sample. As the diffraction pattern shows, the sample is composed of an amorphous and respectively a crystallized phase. The crystallized phase of the sample is a mixture of complex oxides: copper zinc iron oxide, periclase, willemite, diopside, hematite and probably a small amount of others.

Figure 2 b) shows the compositional analysis of the investigated sample determined by EDAX analysis.



a) b)

Fig. 2 – (a) X-ray diffraction spectrum, for powder ash investigated sample;
(b) the EDAX quantification of the sample.

The EDAX analysis indicates a high content of carbon in the investigated sample, where $W_t(\%)$ represents the mass fraction and $A_t(\%)$ is the atomic fraction, in percents.

The measurements of the complex dielectric permittivity over the frequency range 25 Hz to 2 MHz were performed using an RLC-meter Agilent type (E4980A). The ash powder sample was introduced into a cylindrical capacitor, with volume $V = 15.6\,\text{cm}^3$ which is connected to the RLC-meter. For a constant temperature, both the capacitance and quality factor were measured, in the presence of the sample (C_p, Q_p), as well as in its absence (C_0, Q_0), over the working frequency range. The real and imaginary components (ε' and ε'') of the complex dielectric permittivity were determined using the following relations [9]:

$$\varepsilon' = \frac{C_p}{C_0}; \qquad \varepsilon'' = \frac{Q_0 C_p - Q_p C_0}{Q_p Q_0 C_0} \tag{7}$$

3. RESULTS AND DISCUSSION

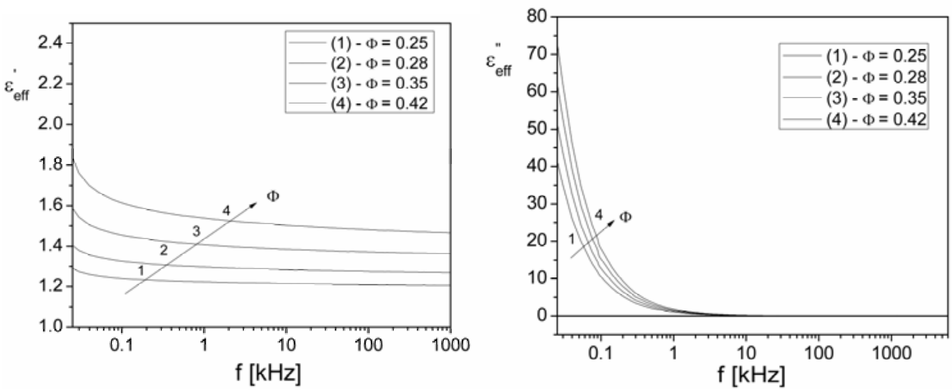
The complex dielectric permittivity of the ash powder sample (a mixture containing micro/nanoparticles dispersed in air), obtained from the dielectric measurements, is an effective complex dielectric permittivity [7], given by:

$$\tilde{\varepsilon}_{eff} = \varepsilon'_{eff} - i\varepsilon''_{eff} \tag{8}$$

In Eq. (8) ε'_{eff} and, respectively ε''_{eff} are the real part and the imaginary part of the effective complex dielectric permittivity, which respects a Debye type dependence, similar to the ones in equations (3) and (4).

The experimental frequency dependence of the real ε'_{eff}, and imaginary ε''_{eff} components of the effective complex dielectric permittivity $\tilde{\varepsilon}_{eff}$, of the investigated ash powder sample, dispersed in air, for different volume fractions Φ of particles in the mixture, are presented in figure 3.

One can observe from figure 3 a) that for each volume fraction Φ, the real part of the effective dielectric permittivity ε'_{eff}, decreases with frequency, from the approximate value 1.8 to the approximate value 1.2. The imaginary component of the effective complex dielectric permittivity, ε''_{eff} decreases with frequency for each constant volume fraction Φ, from the large values to approximately zero (figure 3 b)). The large values of ε''_{eff} at the beginning of the measurement frequency range, are an indication of high conduction losses of the sample (see fig. 3 b)) [7], this fact being in accordance with the EDAX analysis (see fig. 2 b)), which indicates a high content of carbon in the investigated sample.



(a) (b)

Fig. 3 – The frequency dependence of the measured real ε'_{eff}, and imaginary ε''_{eff} components of the effective complex dielectric permittivity $\tilde{\varepsilon}_{eff}$, of the sample for different volume fractions Φ of particles in the mixture

Also, over the entire frequency range, both ε'_{eff} and ε''_{eff}, increase with volume fraction Φ, for a constant frequency. For the effective dielectric constant of the composite systems, there have been developed many approaches involving theoretical studies [10, 11] and also experimental studies [9, 12, 13]. The volume-fraction average model [14] is a simple method estimating the effective dielectric constant, of a mixture/composite system containing two phases:

$$\varepsilon_{eff} = (1-\Phi)\varepsilon_m + \Phi\varepsilon_p \tag{9}$$

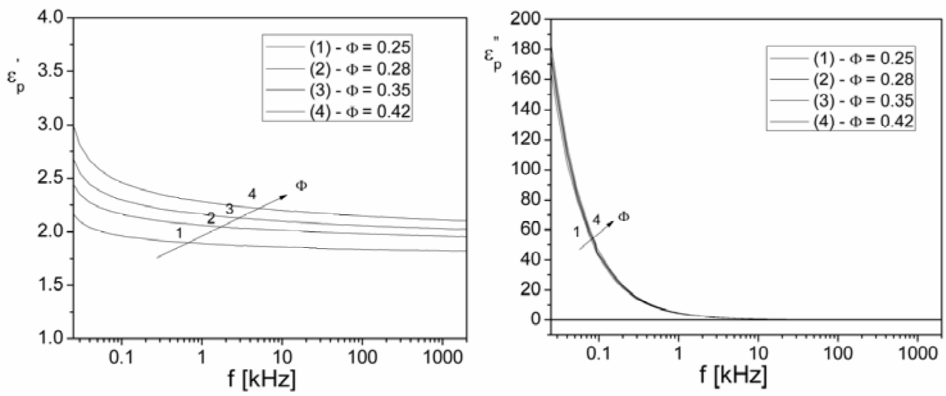
where Φ is the volume fraction of the particles from mixture.

For the investigated ash powder sample dispersed in air, using the complex form, the dielectric constants in Eq. (9), the real ε'_p and the imaginary ε''_p, components of the particle, were computed with equation (10). The dispersion medium being the air, we have used in Eq. (9) the following values for the components of complex dielectric permittivity: $\varepsilon'_m(air) = 1$ and $\varepsilon''_m(air) = 0$. We obtain:

$$\varepsilon'_p = \frac{\varepsilon'_{eff} - 1 + \Phi}{\Phi}, \qquad \varepsilon''_p = \frac{\varepsilon''_{eff}}{\Phi} \tag{10}$$

The frequency dependence of the ε'_p and ε''_p components of the effective complex dielectric permittivity of the ash particles, computed with Eqs. (10), for different volume fractions Φ, are presented in figure 4.

One can observe from figure 4 a) that for each volume fraction Φ, the real part of the ash particle dielectric permittivity ε'_p, decreases with frequency, from the approximate value 3 to the approximate value 2. The imaginary component of the dielectric permittivity ash particle ε''_p, has approximately the same value for all volume fractions Φ, and decreases with frequency, from the large values to approximate zero (figure 4 b)). The large values of ε''_p from the beginning of the measurement frequency range, are an indication of the high conduction losses in sample (see fig. 4 b)), this fact being in accordance with the frequency dependence of the imaginary component ε''_{eff}, of the effective dielectric permittivity (see fig. 3).



(a) (b)

Fig. 4 – The frequency dependence of the calculated real (ε'_p), and imaginary (ε''_p) components of the complex dielectric permittivity of the ash particles for different volume fractions Φ of ash particles dispersed in air.

By introducing in Eqs. (5) and (6) the calculated values for ε'_p and ε''_p (figure 4), and considering that the dispersion medium is the air (with $\varepsilon'_{air} = 1$ and $\varepsilon''_{air} = 0$), we can determine the real part, $\mathrm{Re}\left[\tilde{K}(\omega)\right]$ and imaginary part, $\mathrm{Im}\left[\tilde{K}(\omega)\right]$ of the Clausius-Mossotti complex factor. The frequency dependence of the $\mathrm{Re}\left[\tilde{K}(\omega)\right]$ and $\mathrm{Im}\left[\tilde{K}(\omega)\right]$ parts of the CM factor is shown in Figure 5.

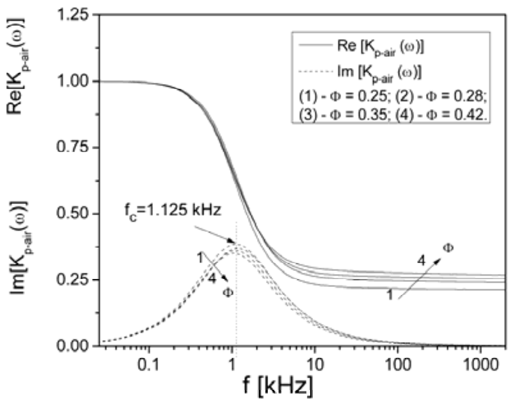


Fig. 5 – The frequency dependence of the real $\mathrm{Re}[\tilde{K}_{p-air}(\omega)]$ and imaginary $\mathrm{Im}[\tilde{K}_{p-air}(\omega)]$ components of the Clausius-Mossotti factor, for different volume fractions Φ of the particles from powder sample dispersed in air.

As seen in figure 5, the frequency dependence of the real and imaginary components of the CM factor fits with a Debye type dependence [7]. The real part of CM factor ($\mathrm{Re}\left[\tilde{K}_{p-air}(\omega)\right]$), for all investigated frequency range is positive. Therefore, dielectrophoresis is positive (pDEP) and, in this case it can do a filtering of flue gas using pDEP, by trapping the nanoparticle in the area of the strongest electric field. The imaginary component of the CM factor, $\mathrm{Im}\left[\tilde{K}_{p-air}(\omega)\right]$ presents a maximum at the same frequency, $f_c = 1.125\,\mathrm{kHz}$, regardless of the volume fraction Φ, of ash particles dispersed in air. The associated peak of the imaginary component, at the frequency f_c, (fig. 5), is correlated with the relaxation time, τ_{MW}, by the Debye relation, $2\pi f_c \tau_{MW} = 1$ [6]. The value obtained for the relaxation time is $\tau_{MW} = 0.1414\,\mathrm{ms}$ and this relaxation time is called dipolar Maxwell-Wagner relaxation time, being typical for the particles with dielectric and conduction losses [7], suspended in a similar medium, with important applications in dielectrophoretic manipulation of nanoparticles or bio-particles and macromolecules [15].

Because the CM factor represents a measure of the relative permittivity between the particle and the surrounding medium, the real component, $\mathrm{Re}\left[\tilde{K}(\omega)\right]$ determines the sign of the DEP force. We have theoretically analyzed the sign of the real component of CM factor when the ash particles are dispersed in a medium without loss, whose dielectric permittivity ε_m, varies from 3 to 10. We have determined in both cases, the real $\mathrm{Re}\left[\tilde{K}_{p-m}(\omega)\right]$ and imaginary $\mathrm{Im}\left[\tilde{K}_{p-m}(\omega)\right]$ components of the Clausius-Mossotti factor in Eqs. (5) and (6) using the calculated values of ε'_p and ε''_p (figure 4), and considering two cases for the surrounding medium: 1) $\varepsilon'_{m,1}=3$ and $\varepsilon''_{m,1}=0$; 2) $\varepsilon'_{m,2}=10$ and $\varepsilon''_{m,2}=0$. Figure 6 shows the frequency dependence of these components, for two values of the volume fraction of the ash particles dispersed in a fluid medium ($\Phi=0.25$ and $\Phi=0.42$).

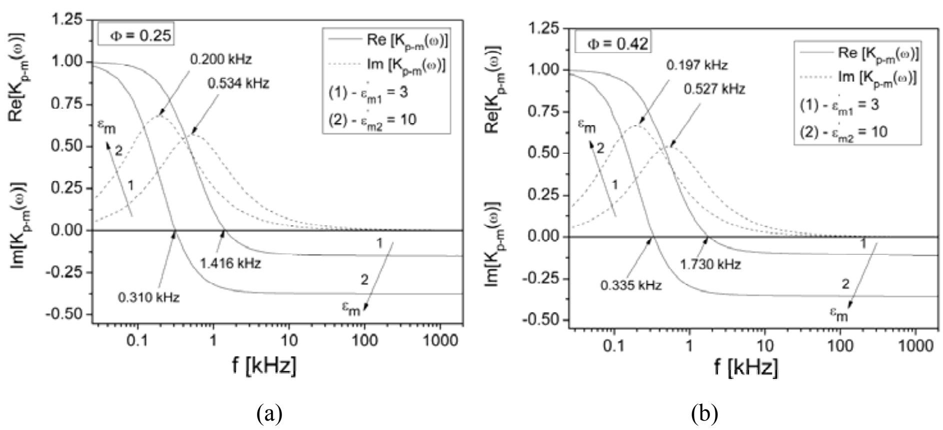


Fig. 6 – The frequency dependence of the real $\mathrm{Re}[\tilde{K}_{p-m}(\omega)]$ and imaginary $\mathrm{Im}[\tilde{K}_{p-m}(\omega)]$ components of the Clausius-Mossotti factor, for volume fraction of the particles, $\Phi = 0.25$ (a) and $\Phi = 0.42$ (b), dispersed in different media with $\varepsilon_m \in [3,10]$.

As seen in figure 6, the real component $\mathrm{Re}[\tilde{K}_{p-m}(\omega)]$, of the CM factor changes its sign, from positives values to negatives values for both analyzed volume fractions of the ash particles dispersed in any fluid medium having a dielectric permittivity, $3 \le \varepsilon_m \le 10$. The sign change occurs at a critical frequency f_c, which decreases from 1.416 kHz to 0.31 kHz (for $\Phi=0.25$) and from 1.730 kHz to 0.335 kHz (for $\Phi=0.42$), if the dielectric permittivity of the medium increases from 3 to 10. Therefore, there is a shift from positive dielectrophoresis (pDEP) to negative dielectrophoresis (nDEP), and the particles can move from regions of high electric field gradient to regions of low electric field gradient. This result shows that it's possible to use nDEP in competition with pDEP to a selective separation of nanoparticle, depending on the frequency of the electric field, on the dispersion medium and the physical properties of ash nanoparticles.

4. CONCLUSIONS

The frequency dependence of the real ($\mathrm{Re}[K(\omega)]$) and imaginary ($\mathrm{Im}[K(\omega)]$) components, of the Clausius-Mossotti (CM) factor were determined based on dielectric measurements of the real, $\varepsilon'(\omega)$ and imaginary, $\varepsilon''(\omega)$ components of the complex dielectric permittivity in the range 25 Hz – 2 MHz, for an ash powder sample resulted from combustion processes and taken from the flue gas filters of a municipal waste incineration plant.

The results show that if the ash powder is dispersed in air, $\mathrm{Re}[\tilde{K}(\omega)] > 0$, for all frequencies in the investigated range. Therefore, a filtering of flue gas is possible, using positive dielectrophoresis (pDEP), by trapping the nanoparticle in the area of the strongest electric field.

The sign of the real component of CM factor, was theoretically analyzed when the ash particles are dispersed in a medium without loss, whose dielectric permittivity ε_m, varies from 3 to 10. The results show that the real component $\mathrm{Re}[\tilde{K}_{p-m}(\omega)]$, of the CM factor changes its sign, from positives values to negatives values at a critical frequency f_c, which decreases from 1.416 kHz to 0.31 kHz, if the ε_m increases from 3 to 10.

The sign change of the real component of the CM factor, shows that there is a shift from positive dielectrophoresis (pDEP) to negative dielectrophoresis (nDEP), and the particles can move from regions of high electric field gradient, to regions of low electric field gradient. Therefore, it is possible to use nDEP in competition with pDEP to a selective separation of ash nanoparticle, depending on the frequency of the electric field, on the dispersion medium and on the physical properties of ash nanoparticles.

The preliminary results obtained show that, there is the possibility of using the DEP for retaining, through manipulation and spatial separation controlled by the ash powder nanoparticles, leading to the purification of exhausted combustion gases and the reduction of the air pollution.

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