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Combustion assisted plasma-chemical processes and environmental effects

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Abstract

A comprehensive analysis of charged gaseous and particulate species formation in flames and inside the combustors as well as in the exhaust plumes of aero-engines and energetic machines are conducted. Some new experimental and model results which exhibit a role of plasma-chemical processes in the pollutant formation are presented.

Keywords: Plasma-chemical processes; Charged particles; Ions; Combustion

1 Introduction

Burning of hydrocarbon fuels initiates different plasma-chemical processes. A number of investigations were focused on the formation of ions in flames (see, for example [1-6]). Recent studies have demonstrated that besides ions and electrons combustion exhausts may contain positively as well as negatively charged nanoparticles such as carbon clusters and soot aerosols [6-9]. Thus, the combustion exhausts exhibit typical properties of low-temperature dusty plasmas. Apart from the fact that the combustion plasmas have much in common with conventional discharge plasmas they have a number of specific features: absence of external electric field, high rate of chemiionization reactions, which produce ions and electrons, a broad range of sizes of clusters and particles (1-150 nm in diameter) charged both positively and negatively, and a relatively small residence time of gaseous products and particles in the high temperature region.

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A considerable interest to study plasma-chemical processes in combustion plasmas is related to the significant environmental effects due to the emission of combustion exhausts to the atmosphere and to the development of novel approaches to control the pollutant formation. In fact, as is believed, soot particles emitted by aircraft engines and energetic machines can considerably affect the Earth's energy budget, induce the additional contrails and cirrus clouds that lead to climatic changes [10]. Ions and ionic clusters, which are produced inside the combustor and in the exhaust plume, are the ones of the most important combustion products affecting the volatile aerosol formation in the aircraft plume and in the atmosphere [11, 12]. As known, ions can induce a nucleation in a vapour-gas mixture and facilitate the coagulation affecting the size distribution of aerosol particles [11-13]. The other possible mechanism of the ion influence on volatile and non-volatile (with soot core) particle formation is the ion-soot interaction. Ions and electrons may attach to particles and induce a charge on their surface [9]. Absorbability of soot particles, features and rate of particle clustering, morphology of soot aggregates, their optical and heterogeneous properties strongly depend on the charge acquired by particles.

This work is focused on the analysis of plasma-chemical processes during combustion of hydrocarbon fuels and on the possible mechanisms of the influence of charged species in combustion exhausts on the environment.

2 Plasma-chemical processes in flames and inside the combustors

Kinetics of ion formation in flames has been investigated during several decades. Ion composition strongly depends on the fuel/air equivalence ratio, ϕ , and on the type of fuel (hydrogen, hydrocarbons, aviation kerosene, etc.). The most frequently used technique to measure the ion concentration in flame is mass-spectrometry [4]. Numerous types of positive and negative ions such as $C_2H_3O^+$, $C_3H_3^+$, CH_3^+ , HCO^+ , $C_3H_5O^+$, $C_3H_7O^+$, H_3O^+ , H_2O^+ , O_2^+ , NO^+ , NO_2^+ , HCO_2^- , HCO_3^- , CO_3^- , CO_4^- , O_2^- , OH^- , NO_2^- , NO_3^- , CN^- were observed in hydrocarbon/air flames. In fuel rich flames, may form heavy hydrocarbon ions $C_{13}H_9^+$, $C_{19}H_{11}^+$, $C_{22}H_{12}^+$, $C_{55}H_{19}^+$, as well as positively charged fullerene molecules [6]. Measurements behind the aviation combustor demonstrated the presence of $C_xH_yO^+$ and $C_xH_yO^-$ ions as well as SO_3^- , SO_4^- , and HSO_4^- ions [14].

In order to understand the mechanisms and principal pathways of ion formation the experimental data should be supplemented by a numerical simulation. In this light, rather complicated kinetic models of charged species formation during combustion of various fuels have been developed [3, 5, 15]. The typical computed spatial profiles of ion mole fractions and temperature for stoichiometric

propane/air flame is shown in Fig. 1. One can find that positive $C_2H_3O^+$ ions and negative HCO_2^- ions as well as electrons prevail in the flame front. Slightly behind the flame front, the H_3O^+ becomes the main ion in the flame due to fast decay of $C_2H_3O^+$. The electrons remain the most abundant negatively charged species behind the flame front. The HCO_3^- is found to be the main negative ion in this flame region.

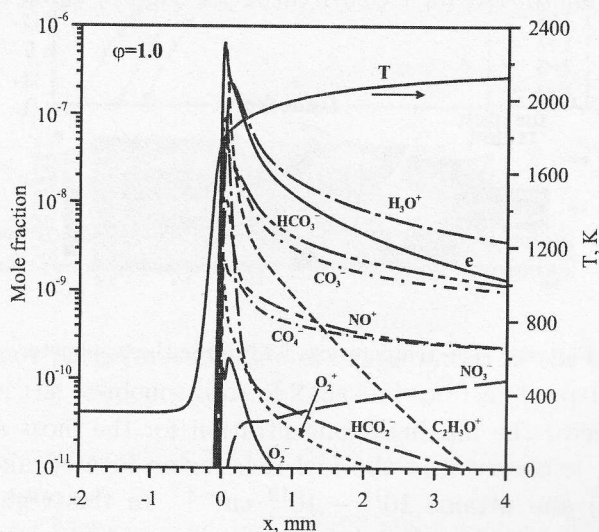


Figure 1. Predicted evolution of ion mole fractions and temperature along a stoichiometric propane/air atmospheric pressure flame.

During combustion of complex hydrocarbon fuels, for example, kerosene containing sulphur in a trace amount, besides these ions the S-containing charged species such as SO_4^- , SO_3^- , HSO_4^- form. Depending on the value of ϕ SO_3^- and HSO_4^- may be the most abundant negative ions behind the flame front [9, 15].

The processes of ion formation inside the aero-engine or energetic machine combustors are more complicated than those in flames. The modern combustors operate in a diffusion mode. In this case, the fuel/air mixture composition differs considerably in various regions of combustor. Therefore, a number of different ions can form inside the combustor. The gas residence time for combustors (5-10 ms) is much smaller than that for flames (~ 100 ms). The total ion concentration of $\sim 2 \cdot 10^8 \text{ cm}^{-3}$ at the exit plane of modern aero-engine combustor was measured recently by Haverkamp et al. [16]. However, in present, the concentration of ions and electrons inside the combustor may be estimated only by computations.

To simulate the processes of charged gaseous species formation inside an aviation combustor one have to apply approximate combustion models, for example, the Flame Let model [17]. In our computations this model was supplemented by

the ion kinetics developed in [15]. To calculate the flow parameters inside the combustor operating in a diffusion mode (these parameters are needed as input ones for the Flame Let model) with outlet pressure of 10^6 Pa and temperature $T_c = 1540$ K the three-dimensional numerical simulation of turbulent combustion using the standard STAR-CD software with the equilibrium chemistry was conducted. Figure 2 shows the temperature field inside the typical combustor (this combustor is similar to that for PC90A turbo-jet engine) using the methane as a fuel.

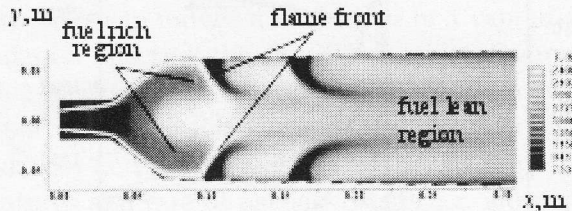


Figure 2. The temperature field inside the combustor.

Inside the combustor the maximal concentration for the most abundant ions in flames, $C_2H_3O^+$, is observed in the fuel rich region located ahead of the flame front (see Fig. 2) and attains $10^{11} - 10^{12} \text{ cm}^{-3}$. In this region, the maximal concentration for negatively charged species is appeared for electrons and HCO_2^- ions. However, at the combustor exit the concentrations of these species are negligible. In the fuel lean zone of combustor at the exit plane only NO^+ , H_3O^+ , SO_3^- , HSO_4^- or NO_3^- ions remain to be abundant. The predicted total concentration of positive (negative) ions in the combustor outlet is around $2 \cdot 10^8 \text{ cm}^{-3}$ that is in a good agreement with measurements [16]. The ion composition strongly depends on the value of fuel sulphur content (FSC). The FSC can vary from 0.001% to 0.3% (per mass) for modern aviation fuels.

Recent studies indicated that engine-generated soot has a rather high conductivity [18]. Hence, the soot particles turn out to be polarized in the ion-produced electric field and to compute the ion-soot attachment coefficient one should take into account the exerting both Coulomb and image forces. The ion-soot attachment coefficient depends on temperature, ion mass, particle size, ion and particle polarities [9]. Figure 3 plots the dependence of the attachment coefficient for NO^+ ion, $\beta^{(P)}$, on the radius of soot particle, a , with different charge (parameter P is the product of the ion and particle charges). One can see that ions can be attracted to the particles with the same polarities. Even for small size particle with radius $a = 5$ nm the probability to accumulate the charge larger than unity is appeared. This means that clusters and soot particles in flame and inside a combustor may acquire a rather large charge. Figure 4 depicts the predicted charge

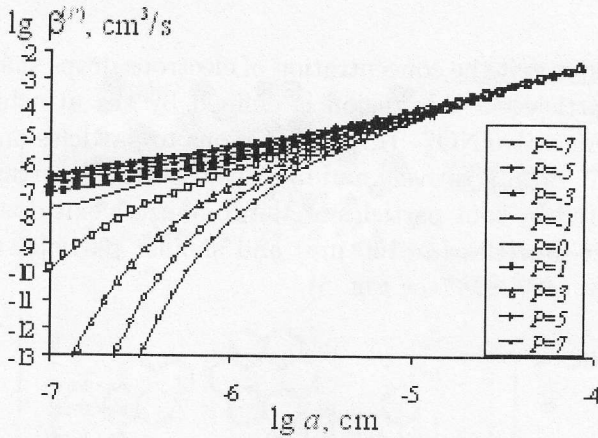


Figure 3. Attachment coefficient for NO^+ ions, $\beta^{(P)}$, vs. particle radius, a , for different values of the parameter P at temperature $T = 2000$ K and pressure $p = 0.1$ MPa.

distribution of soot particles with different radii ahead of the flame front inside the combustor for the residence time of 2 ms and total soot particle concentration $N_s = 10^7 \text{ cm}^{-3}$.

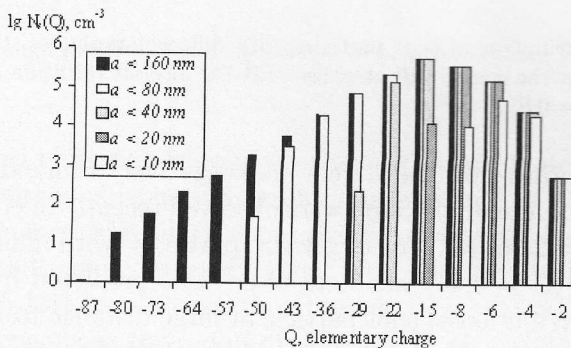


Figure 4. Charge distribution of soot particles with radius, a , smaller than given value in the reactor corresponding to the fuel rich region ahead of the flame front inside the combustor with $T_c = 2200$ K, $p_c = 1$ MPa ($\phi = 2$).

Prediction of the dynamics of soot particle charging was conducted by using the approach developed in [9]. Size distribution of soot particles was assumed to be log-normal with medium radius of 25 nm and geometrical deviation of 1.56. Attachment of electrons to particles in the fuel rich region of combustor leads to formation of negatively charged ensemble of soot particles in which the large size particles (with radius $a \geq 80$ nm) can acquire the charge $Q = 40 - 70e$ (e is an elementary charge) and small size particles with $a = 10$ nm may accumulate the

charge $Q \leq 8e$.

To the combustor exit the concentration of electrons drops sharply. The charge distribution of particles in this region is defined by the attachment of positive NO^+ , H_3O^+ and negative NO_3^- , HSO_4^- , SO_3^- ions to particles and becomes to be more symmetrical. Both positively and negatively charged particles are observed. The charge acquired by soot particles at the combustor exit does not exceed $10e$ even for large size particles ($a=160$ nm) and smaller particles with $a = 10$ nm acquire the charge $Q = \pm 2e$ (see Fig. 5).

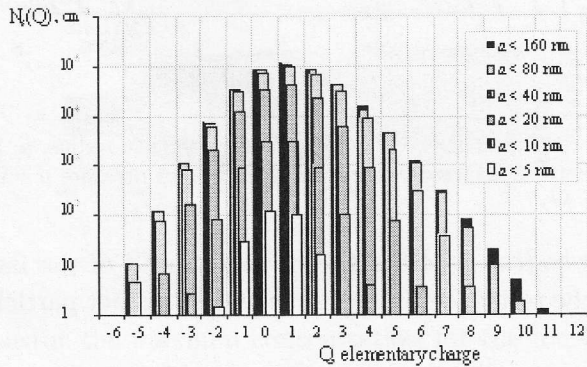


Figure 5. Charged distribution of soot particles with different radius, a , that is smaller than given value at the engine exit at cruise of B-747 aircraft (altitude $H = 10.7$ km, Mach number $M_0 = 0.8$).

It is worth to note that both positively and negatively charged up to $Q = \pm 2e$ soot particles with diameter of 13 nm were observed recently in ethylene/air atmospheric flame at $\phi = 2.06$ [19]. For larger size particles (with diameter $d = 24$ nm) the greater charge ($Q = \pm 4e$) was detected. As is predicted by our model, the detected charge distribution of soot particle at large distance from flame front appeared to be symmetrical, however, slightly behind the flame front the density of negatively charged particle is considerably larger than that for positively charged ones.

As is believed, the soot particles form from polyene molecules and polycyclic aromatic hydrocarbons (PAH) [20]. Figure 6 shows the principal scheme of soot particle formation presented in [20].

In accordance to modern models, the pyrene molecules which consist of four aromatic rings are the main soot precursors. The particle inception with radius of approximately 1 nm is constructed from PAH. In the fuel rich zone of combustor the concentration of pyrene molecules can be as large as $10^{12} - 10^{14} \text{ cm}^{-3}$ and concentration of primary clusters (particle inception) attains $10^{11} - 10^{12} \text{ cm}^{-3}$. Due to attachment of electrons and ions to primary clusters the latter ones acquire

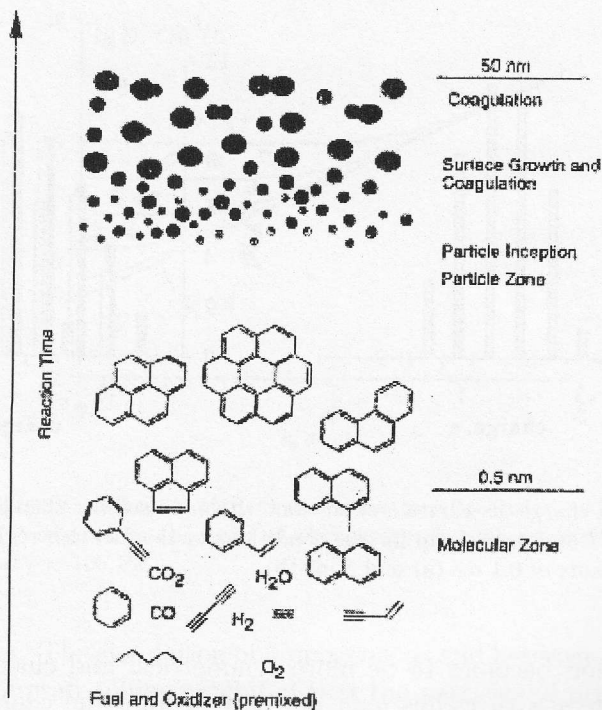


Figure 6. A rough picture for soot formation in premixed flames (Bockhorn, 1994).

a charge. Charged clusters with opposite polarities coagulate more rapidly than neutral ones. Charged clusters induce the image charge on the neutral particles and, as a consequence, the image attractive force appears between charged and neutral clusters that enhances coagulation. The calculations based on the procedure presented in [21] have shown that the enhancement factor for coagulation rate coefficient depends on the radius of the particle, on the cluster charge, and on temperature. At $T_0 = 2000$ K for neutral and singly charged clusters this factor may achieve 2. For cluster with larger charge ($Q = 3e$) and $a = 1$ nm it may be as large as 4. Figure 7 depicts the predicted charge distribution of clusters with different sizes formed in the fuel rich ($\phi = 3$) zone of combustor from the primarily neutral monodisperse precursors with $a_0 = 1$ nm at their initial concentration $N_{c0} = 10^{11}$ cm $^{-3}$ for time instants $t = 0.1$ and 5 ms. The calculation model took into account charging of clusters in the fuel rich region of combustor and coagulation growth due to exerting of Coulomb, image, and Van der Waals forces between clusters.

As is seen, at an initial stage even small size clusters are principally charged negatively due to the strong electron attachment. At the latter time instants the

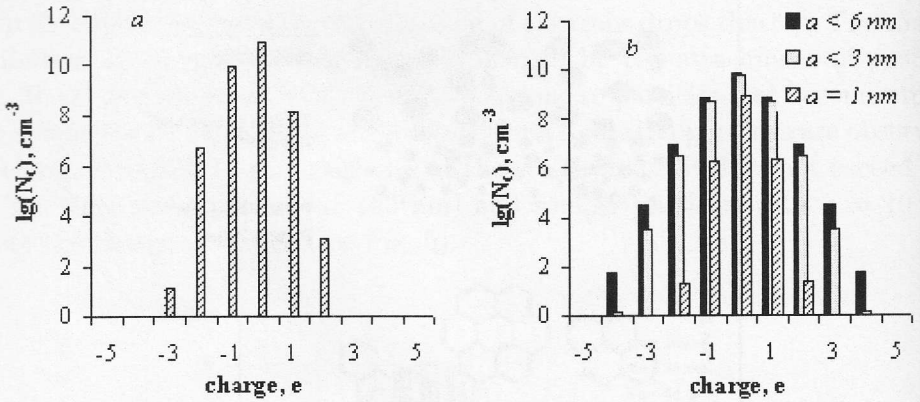


Figure 7. Predicted charge distribution of clusters with radii smaller than given value at $N_{c0} = 10^{11} \text{ cm}^{-3}$ and $a_0 = 1 \text{ nm}$ for the conditions in the fuel rich region of combustor for time instants of 0.1 ms (a) and 5 ms (b).

charge distribution becomes to be more symmetrical and cluster sizes increase. Even small clusters with radius $a=4 \text{ nm}$ may acquire the charge $Q = 4e$. It is worth to note that accounting only the Coulomb interaction between oppositely charged clusters does not allow one to form large clusters with $a = 6 \text{ nm}$ for time interval of 5 ms due to coagulation process.

The estimations exhibited that a marked amount of polar molecules, mostly H_2O , SO_2 , SO_3 , HNO_2 , may be acquired by charged soot particles or charged clusters in aviation combustor for residence time $\tau_{res} \sim 5 - 10 \text{ ms}$. This means that noticeable proportion of soot particles ($\sim 10 - 20\%$) may be activated to act as contrail nuclei directly inside the combustor. Measurements demonstrated that combustor-generated soot particles consist of two different fractions. One of them was found to contain the impurities of a number of sulphates, nitrates, and organics and exhibits a high level of hydrophilicity. This fraction is supposed to form from particles with a large charge, $Q > 10e$, or from relatively small clusters ($a \approx 1 \text{ nm}$) consisting of polyaromatic hydrocarbon molecules forming primarily in the fuel rich region of combustor and having a small charge, $Q = 1 - 3e$. The attachment coefficient of polar molecules to charged clusters is by a factor of 20-50 higher as compared to neutral ones. This is illustrated in Fig. 8, which shows the dependence of attachment coefficient for H_2O molecule to soot particle with various charge, Q , on particle radius.

The other fraction was found to be hydrophobic and is supposed to form from neutral clusters and particles or larger particles ($a > 10 \text{ nm}$) with small charge, $Q \leq 2e$. These particles can not accumulate polar molecules during residence in-

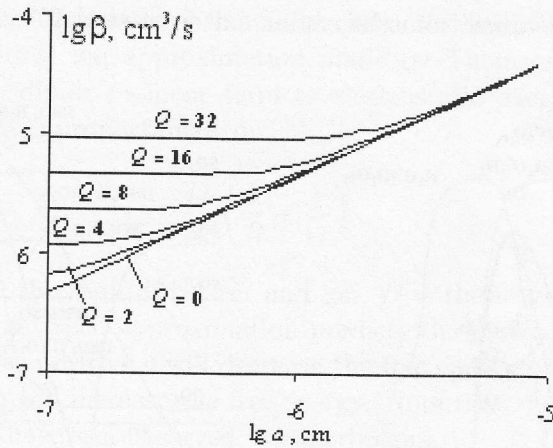


Figure 8. The attachment coefficient of H_2O molecules to soot particle with different charge value, Q (Q is given in the units of elementary charge), vs particle radius for $T = 2000$ K and $p = 106$ Pa.

side the combustor. The simulation of homogeneous and heterogeneous nucleation processes in the aircraft plume exhibited that the particles of primary hydrophobic fraction are partly activated to uptake the water molecules during the normal condensation process due to deposition of the soluble matter ($\text{H}_2\text{O} / \text{H}_2\text{SO}_4$) on their surface. The binary heterogeneous nucleation on the surface of soot particles and coagulation of the emitted soot particles with sulfate volatile aerosols were found to be responsible for activation of soot hydrophobic fraction. It turned out that small size particles with $d < 15$ nm are activated, generally, due to coagulation process and larger particles ($d > 15$ nm) are activated due to binary heterogeneous nucleation. Depending on the fuel sulfur content, 5-20 % particles of hydrophobic fraction may be activated in the plume in these processes.

3 Plasma-chemical processes in the exhaust plume

The other pathway of soot particle activation deals with the existence of charged clusters in the plume. Modelling and experimental studies have shown that various ionic clusters such as $\text{HSO}_4^-(\text{H}_2\text{SO}_4)_m$, $\text{HSO}_4^-(\text{HNO}_3)_{m1}$ ($m = 1..3$), $\text{NO}_3^-(\text{HNO}_3)_q$ ($q = 1..5$), $\text{NO}_3(\text{HNO}_3)(\text{H}_2\text{O})$, $\text{H}_3\text{O}^+(\text{CH}_2\text{O})(\text{H}_2\text{O})_m$, and $\text{H}_3\text{O}^+(\text{H}_2\text{O})_n$ ($n = 1 - 6$) can form in the plume [22, 23]. Figure 9 illustrates the evolution of positive and negative ionic clusters along the plume of B-747 aircraft at cruise ($H = 10.7$ km, $M_0 = 0.8$). One can see that the most abundant charged clusters in the aircraft plume are the hydrated ions with H_3O^+ core and $\text{HSO}_4^-(\text{H}_2\text{SO}_4)_m$ ions. The concentration of $\text{H}_3\text{O}^+(\text{H}_2\text{O})_6$ and $\text{HSO}_4^-(\text{H}_2\text{SO}_4)_3$

clusters at 10 m distance from the engine exit is around 10^6 cm^3 .

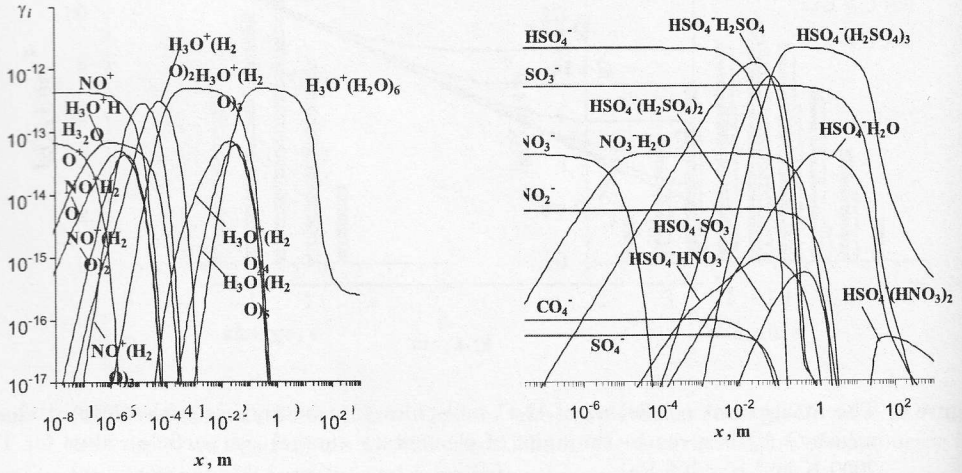


Figure 9. Evolution of ionic cluster mole fractions along the plume axis of B-747 aircraft at cruise for $FSC = 0.04\%$.

Because the ion composition at the engine exit was calculated taking into account the charging of soot particles inside the engine the predicted density of lighter H_3O^+ ions was smaller than that for HSO_4^- ions and, as a consequence, the concentration of ionic clusters with H_3O^+ core is markedly smaller than that for heavy clusters $\text{HSO}_4^-(\text{H}_2\text{SO}_4)_n$. Due to dilution of the plume and attachment of these ionic clusters to soot particles their concentration drops considerably: at 50 m distance the density of these clusters is as small as 10^4 cm^{-3} . It is worth noting that despite the large density of NO^+ ions at the engine exit there are no ionic clusters with NO^+ core at 10 m distance from the engine exit. Such behavior is explained by the fast decay of $\text{NO}^+(\text{H}_2\text{O})_3$ clusters due to reaction $\text{NO}^+(\text{H}_2\text{O})_3 + \text{H}_2\text{O} = \text{H}_3\text{O}^+(\text{H}_2\text{O})_2 + \text{HNO}_2$ [22]. That is why one cannot observe ionic clusters with NO^+ core in the aircraft plumes. The attachment of charged clusters to soot particles changes the particle charge distribution in the plume. Nevertheless, the significant charge continues to be on the particle surface.

Ionic clusters initiate the formation of charged sulfate volatile aerosols via ion-induced nucleation and attachment of charged clusters to neutral sulfate particles caused by exerting of image force. Thus, there exist differently charged aerosols in the plume: ionic clusters ($d < 1 \text{ nm}$), charged volatile sulfate aerosol ($d = 1 - 10 \text{ nm}$), and charged soot particles ($d = 10 - 150 \text{ nm}$). It should be noted that the electrostatic contribution to the free energy of cluster formation is due to attractive force the ion exerts on the molecules in the cluster. This force reduces the free energy of formation for charged clusters as compared to uncharged ones [13]. In ion-induced nucleation the contribution of electrostatic field to the

free energy of cluster formation is not fully characterized by the ion charge, as in the classic spherical ion approximation made by Thomson. The important contribution of the dipole moment term to electrostatic energy may come into play [24]. This term is proportional to

$$\frac{D}{2q} \left(\frac{1}{R} + \frac{1}{r_i} \right),$$

where R and r_i are the radii of cluster and ion, D is the ion dipole momentum, q is the charge of ion. Such approximation predicts the preference for nucleation of positive ions, since positive q will decrease the free energy of formation and, in contrast, negative q will increase the free energy. Note that the ion-sign effect on the nucleation rate has been observed in experiments.

Heterogeneous binary $\text{H}_2\text{O}/\text{H}_2\text{SO}_4$ nucleation on the surface of charged soot particles occurs much faster than on the neutral particle one. This process is responsible for the additional deposition of soluble matter on the soot particle surface and, thus, increasing the number of activated particles. Charged particles with opposite polarities coagulate much faster than neutral ones. Moreover, the coagulation between charged and neutral particles occurs also significantly faster due to appearance of electric image force. The enhancement factor for coagulation coefficient, in this case, as compared to coagulation of neutral particles depends on particle sizes and may be as large as 20 even for the interaction of neutral and singly charged particles. The characteristic coagulation time for charged-charged and charged-neutral particle interaction under plume conditions varies from 0.1 to 1 s that is smaller than the gas residence time in the plume, $\tau_{res} \approx 2-4$ s. The coagulation between such particles in the plume may lead to formation of large size aggregates with $d \geq 150$ nm, which can act as a cloud condensation nuclei. The appearance of a charge on the aerosol particle surface can accelerate the normal condensation process and lower the saturation value required to vapor-liquid phase transfer due to a decrease of Kelvin barrier caused by attraction of dipole molecules such as H_2O , H_2SO_4 , HNO_3 , organics by charged droplet and, thus, can facilitate the contrail formation.

4 Concluding remarks

A number of plasma-chemical processes proceed during combustion of hydrocarbon fuels. The combustion plasmas exhibit the typical properties of low temperature dusty plasmas: the presence of charged gaseous and particulate components, the complex interaction between ion (electrons) and cluster systems, significant influence of particle and cluster charging on the heterogeneous processes and coagulation of clusters. Combustion-assisted plasma-chemical processes can affect

considerably the pollutant formation and contribute the environmental effects. To summarize one can conclude that the abundance of clusters and particles having a significant electrical charge inside the combustor and in the combustion exhaust may be used for scavenging of small size volatile aerosols and molecules with permanent dipole momentum and, thus, to control the pollutant emissions.

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