Agglomeration processes in carbonaceous dusty plasmas, experiments and numerical simulations

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Abstract. This paper deals with carbon dust agglomeration in radio frequency acetylene/argon plasma. Two studies, an experimental and a numerical one, were carried out to model dust formation mechanisms. Firstly, in situ transmission spectroscopy of dust clouds in the visible range was performed in order to observe the main features of the agglomeration process of the produced carbonaceous dust. Secondly, numerical simulation tools dedicated to understanding the achieved experiments were developed. A first model was used for the discretization of the continuous population balance equations that characterize the dust agglomeration process. The second model is based on a Monte Carlo ray-tracing code coupled to a Mie theory calculation of dust absorption and scattering parameters. These two simulation tools were used together in order to numerically predict the light transmissivity through a dusty plasma and make comparisons with experiments.

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1. Introduction

In the frame of the future International Thermonuclear Experimental Reactor (ITER), several feasibility studies were launched not only to design the reactor, but also to define the operating conditions and the security parameters. Related to this last issue, a dedicated topic deals with the 'plasma-facing components' and more generally about the plasma–wall interaction. High-energetic fluxes are expected in some areas, for example in the divertor zone of a tokamak where important erosion and dust production have already been observed [1]. The dust occurrence in fusion plasma induces several drawbacks. On the one hand, there are security issues: firstly, dust particles in tokamak systems may retain a large fraction of isotopic hydrogen as tritium and incidentally could spark off major safety issues [2, 3]. Secondly, these finely dispersed particles with high chemical reactivity may spontaneously react with any oxygen or water vapor in the presence of vacuum or coolant leaks. Thirdly, radioactive dust particles migrate inside the tokamak under the influence of many physical forces [4] and may concentrate at the colder reactor areas through repetitive evaporation and condensation. Consequently, the heat transfer to actively cooled components may be hindered by the thermal resistance of the deposited dust layers. On the other hand, a plasma charged with dust particles loses some reactor power and the fusion device efficiency can be seriously lowered, especially when dust is made of high-atomic-number species. Thus, understanding dust formation, aggregation and transport in plasma should be achieved in order to limit or to foresee the previously exposed problems. Besides, there are several other research areas concerned with dusty plasma. For example, in the semiconductor industry, dust occurrence during plasma processing is often considered as a major problem and several studies have been carried out in this field [5]. Another example concerns astrophysics and especially interstellar clouds and planetary atmospheres, which contain dust that can be produced with plasma discharges [6, 7].
The observation of plasma dust, obtained by wall erosion in a tokamak, can be achieved through \textit{ex situ} analysis of the dust deposited on the reactor parts as was often done. Another technique is \textit{in situ} analysis of the dust present in the plasma during fusion operation. For such an approach, optical diagnostics are particularly interesting. For example, particle motion can be observed with a fast visible camera \cite{8}. The emitted light of the radiating species is detected and path lines can be identified showing the main deposition locations. Another optical diagnostic device is the infrared camera, which can be used to monitor dust deposition on some specific surface of the reactor, such as the limiter in Tore Supra experiments \cite{9, 10}. A lot of information can be obtained from these two optical diagnostics. Nevertheless, both of them require specific data post-treatment, which can be long and tricky, to identify the key parameters of dust (namely the size distribution and the number density of dust particles).

One promising technique to obtain efficient dust diagnostic in a tokamak is based on light scattering measurements, similarly to what has been done for aerosol characterization. In the electromagnetic theory, wave propagation in complex media is clearly described and a specific theory of light scattering by spherical particles has been established (Rayleigh and Mie scatterings) as a function of particle size and concentration, wavelength and refractive index. However, the dust produced in a tokamak can be very heterogeneous with a broad range of size, shape and material components. To get rid of uncontrollable parameters such as the dust composition, at the laboratory scale dusty plasma can be made in a capacitively coupled parallel plate reactor. This is the choice that was made in the present study in order to observe and model the dust agglomeration process in a radio frequency plasma even though this plasma discharge is not dense like a tokamak one. Naturally, the purpose of these experimental and numerical studies is to develop a diagnostic tool for further implementation on a tokamak. Besides, there is another difficulty that is not addressed in the present study and which is relevant in tokamak plasma, the magnetic field occurrence. This last issue will be soon investigated at the laboratory scale with the design of a new electron cyclotron resonance (ECR) reactor. This experimental device will be made up of a diffusion chamber containing a sample holder set inside coils. With this reactor, the magnitude and orientation of the magnetic field will be monitored in order to have operating conditions similar to those encountered in a tokamak.

With the current experimental device, the operating parameters of the discharge are well controlled and experiments can be reproduced. The apparatus will be described in the following section. Besides, a model that can help in understanding the scattering measurements during particle growth in the plasma is necessary. The modeling tools dedicated to agglomeration and transmissivity in carbonaceous dusty plasma are presented in section 3. Then, section 4 gives some comparisons between experiments and simulations. The main mechanisms representative of the dust agglomeration and their incidence on the optical measurement are thus discussed.

2. Experiments

2.1. Reactor and acquisition devices

Experiments were carried out in a cylindrical diode RF reactor (figure 1(a)) with an asymmetric configuration. The reactor vessel is pumped by a rotary pump and the operating pressure is about 200 mTorr (26.66 Pa). Inside the reactor the plasma is generated between the cathode located at the top of the reactor (stainless steel or carbon), which is powered through an impedance
matching network and a grounded electrode at the bottom separated by $h = 8\,\text{cm}$. Their size and shape can be modified according to the experimental requirements.

Several acquisition devices can be implemented around the reactor. On the one hand, electrical measurements such as bias voltage and RF power delivered to the plasma were performed. On the other hand, optical diagnostics dedicated to the carbonaceous dust observation were achieved. Among them, spectroscopic devices (in the visible and infrared ranges) and a camera were used. Infrared spectroscopy was performed using a Bruker FTIR Equinox 55 device. Its working wavelength range is $2.5\,\mu\text{m} \rightarrow 25\,\mu\text{m}$; detection was done with a cooled MCT detector. UV–visible spectroscopy was performed with an Avantes spectrometer (Avaspec 2048) operated between 200 and 1100 nm. This apparatus was used with a deuterium halogen source or with a diode laser ($\lambda = 405$ or $623\,\text{nm}$). Besides, spectroscopic measurements for three different observation angles could be performed simultaneously. Eventually, a Photron Fastcam-SA1 camera was used for video acquisition of the generated and transported dust around the cathode. With 5400 frames per second in full resolution, this camera could be used to observe the larger dust pathlines. In figure 1(b), a typical arrangement of the light sources and detectors is shown.
Thus, during experiments several wavelength ranges of interest were investigated according to the probe light source and the employed detection device. This point is really important because the intensity of scattered light by dusty plasma is strongly dependent on the size and particle density and the measured signal can greatly vary with the detection wavelength. In brief, small particles could be detected in the UV–visible range, larger ones obtained by agglomeration were observed in the infrared range and the biggest ones were followed by the visible camera.

For fast visible camera (CMOS) acquisitions, a specific rectangular cathode was used. With this kind of electrode the carbon dust is confined in a bounded domain that can match the camera focusing plane. In this configuration, the large-particle path can be followed over a longer duration. Data analysis has shown that dust moves along the main dimension of the cathode and vertically oscillates within the plasma sheath [11, 12].

In this study, we only present results concerning small particles; thus UV–visible spectroscopy measurements are the ones that are used to do comparisons with numerical simulation of the agglomeration process.

2.2. Dust production technique

Carbon dust in the reactor chamber can be produced by two distinct methods. The first one, which is quite similar to what happens at tokamak walls, is carbon cathode sputtering. In this case, very small particles are obtained with low concentrations. The sputtering parameters are adjusted using the injected RF power in the plasma, the reactor pressure and the cathode size. The second method, which was the method we mostly used, is based on controlled injection of acetylene into the argon plasma. With this technique, $C_2H_2$ dissociation produces many more carbon precursors than carbon cathode erosion. Therefore, the amount of dust in the plasma promotes agglomeration and biggest particles and higher concentrations are observed. In this case the operating parameters are RF power, reactor pressure and acetylene flow rate. For these experiments, a stainless steel cathode was used. According to the chosen production technique, very different dust mixtures have been observed. Previous works dedicated to dust chemistry and morphology have discussed these results [13].

2.3. Transmissivity and bias measurements

Transmissivity measurement was one of the first diagnostics that was implemented in our experimental device. Previous studies were carried out with an infrared light source in the same reactor in order to monitor large dust particles. Experimental data analyses put forward the fact that agglomeration of small carbon dust particles into larger ones could be assessed from temporal transmissivity variation. In the previous study [14], a bimodal distribution that accounts for the continuous production and agglomeration mechanisms was used to model the optical properties of the dusty plasma. Numerical simulations of light propagation in the corresponding media were in good agreement with experiments. However, infrared probing of plasma dust for size and density characterization is efficient as long as the particles are sufficiently big. In the case of dust diameter smaller than 1 $\mu$m, UV–visible experiments should be more sensitive. The second diagnostic used in our experimental device was bias voltage measurement. When carbon dust is produced, the bias voltage changes since carbonaceous particles collect electrons. In consequence, measurement of bias voltage variations can be a useful test of carbon dust occurrence in the plasma sheath.
Figure 2. Experimental spectra of \( \text{Ar}/\text{C}_2\text{H}_2 \) dusty plasma. Acetylene injection \( t = 180 \) s; gas flow: \( \dot{q}_{\text{Ar}} = 12 \) sccm (cm\(^3\) min\(^{-1}\)), \( \dot{q}_{\text{C}_2\text{H}_2} = 2 \) sccm, \( p_{\text{tot}} = 1.4 \times 10^{-1} \) Torr; maximum injected power \( P_{\text{max}} = 40 \) W. (a) Transmissivity at 400 nm (continuous line), 600 nm (dashed line) and 902 nm (dashed-dotted line). (b) Transmissivity at 500 nm (continuous line); bias voltage (dashed line).

Both types of measurements were carried out for various plasma discharge parameters. Typical transmissivity spectra and bias voltage are shown in figure 2. In this experiment, the positions of the capacitors in the matching network were fixed in order to prevent arc occurrence. Besides, varying capacitances in the matching network induce changes of the bias voltage, which become much more complicated to analyze, because they are not related solely to dust occurrence.

For all experiments, the same operating procedure was applied: halogen source light turned on, argon plasma ignition, acetylene injection, acetylene injection cut-off and argon plasma cut-off. In figure 2(a) transmissivity of the dusty plasma is plotted at three wavelengths: \( \lambda = 400 \), 600 and 902 nm. It can be noted that without acetylene injection \( (t < 180 \) s) the argon plasma is transparent in the UV–visible range. As soon as \( \text{C}_2\text{H}_2 \) is injected into the reactor there is a rapid decrease of transmissivity, especially for small wavelengths for which \( \text{Tr}(400 \text{ nm}) \approx 29\% \) at
At $t = 225$ s, the transmissivities at higher wavelengths are larger: $\text{Tr}(600\text{ nm}) \simeq 45\%$ and $\text{Tr}(902\text{ nm}) \simeq 60\%$. This can be explained by a strong production of fine carbonaceous particles that progressively agglomerate into larger ones. At $t > 225$ s, small particles are more quickly consumed by large dust than they are produced. Thus, transmissivity at high wavelengths keeps decreasing until $t \simeq 385$ s, while it remains almost constant at $\lambda = 400$ nm. Then, transmissivities for all wavelengths increase, probably because of the small dust particles consumption in the plasma sheath until $t = 583$ s. At this stage, there is a full distribution of small and large particles. From this point, periodic fluctuations with an average period of $T = 126$ s can be observed. They are followed by steady-state-like behavior at $t \simeq 2500$ s until acetylene injection is cut off. Other experiments have shown that these transmissivity fluctuations have a duration and a periodicity that strongly depend on the discharge parameters (RF injected power, reactor pressure and gas flow rate). They frequently seem to correspond to dust production cycles (generation, agglomeration and removal).

Concerning the bias voltage (figure 2(b)), similar oscillations with the same period can be observed. The amplitude of the bias signal is correlated with the amount of dust present in the plasma sheath. In the present case, both oscillation patterns are in phase opposition after the transition period ($t > 583$ s). This transition period is very delicate to explain as several mechanisms are involved in the initial bias voltage variation. Among them, there are the strong perturbations in the discharge due to the acetylene injection in the argon plasma that induces important pressure effects. However, after this transient stage when the gas flow becomes steady, the bias voltage variations are mainly related to dust occurrence. As dust is produced, it rapidly captures electrons and the absolute value of the bias voltage decreases. When the transmissivity increases, particle density within the plasma decreases; therefore the absolute value of the bias voltage also increases and a new cycle starts. The bias measurement is a useful diagnostic to detect dust occurrence. However, it does not provide any information about particle size distribution. Actually, there are several scenarios (see e.g. [15]) to explain how electrons are captured and distributed among the created carbonaceous dust.

Considering these experimental data, it is obvious that dust strongly affects light propagation within the plasma. The observed fluctuations of transmitted intensity are correlated with dust production, growth and loss and with sheath fluctuations. In order to achieve an accurate description of the dust size distribution, a model of the agglomeration process is required. The following section provides details of the continuous population balance equations used to model mass conservation of dust within the plasma. Using these calculated size distributions, light transmissivity could be numerically assessed.

### 3. Models and numerical simulations

#### 3.1. Aerosol agglomeration: balance equation

Agglomeration science is an extensive research field with numerous applications. Among several textbooks dealing with particle agglomeration, the early work of Friedlander [16] is a reference. Chapter 7 of this reference deals with aerosol collision and agglomeration that can be modeled by population balance equations. A general formulation of these equations has been reported by Randolph and Larson [17] as

\[
\frac{\partial n(x)}{\partial t} + \nabla \cdot [vn(x)] + \nabla \cdot [wn(x)] = B - D,
\]  

(1)
where \( n(x) \) stands for the distribution of quantity \( x \), which can be either the particle length, volume, surface, mass, etc. The two divergence terms on the lhs of equation (1) are related to spatial variations of the distribution for ‘external’ and ‘internal’ coordinates. The external coordinates are the usual Lagrangian ones that are used to locate a particle in a given control volume; thus \( \mathbf{v} \) is a velocity vector. The internal coordinates are related to spatial variation inside particles due to processes such as shrinkage and growth, \( \mathbf{u} \) being the process speed. In the present work, simplifying assumptions were made. Firstly, particles remain confined in the control volume, i.e. there is no convective transport. Secondly, particles that agglomerate are not subject to any structural volume modification, i.e. two mother particles that agglomerate give birth to one daughter particle whose volume is the sum of the volumes of the mother particles. Therefore, the distribution balance equation is only driven by birth and death source terms \( B \) and \( D \). Besides, the quantity considered is the particle volume \( v \), and the particles are assumed to be spherical.

The birth and death source terms can be defined by integral formulations \[ 18 \], which involve particle density \( n(v) \) and agglomeration kernels \( \beta(v_j, v_k) \)

\[
B(t) = \frac{1}{2} \int_0^v \beta(v', v - v') n(v', t) n(v - v', t) \, dv'
\]

and

\[
D(t) = \int_0^\infty \beta(v', v) n(v', t) n(v, t) \, dv',
\]

where \( \beta(v', v) \) is the probability of agglomerating two particles of volumes \( v \) and \( v' \) during the time step \( dt \).

3.2. Aerosol agglomeration: sectional model

In order to solve the above balance equation, the particle size distribution was handled using a sectional model. With this type of model, the volume range is partitioned into sections \([v_i, v_{i+1}]\). In these sections all the particles have the same representative volume \( x_i \). The density variations of the \( i \)th section \( \frac{\partial N_i(t)}{\partial t} \) correspond to the integration of equation (1) over \([v_i, v_{i+1}]\), i.e. \( N_j(t) \times x_i = \int_{v_i}^{v_{i+1}} N_j(t) \, dv \). The problem is therefore characterized by a set of \( N_{sec} \) differential equations that has been fully described by Kumar and Ramkrishna \[ 19 \]. The population balance is written as

\[
\frac{dN_j(t)}{dt} = \sum_{i,k} \left( 1 - \frac{1}{2} \delta_{j,k} \right) \eta \beta_{j,k} N_j(t) N_k(t) - N_j(t) \sum_{k=1}^{N_{sec}} \beta_{i,k} N_k(t) \quad \text{with} \quad x_{i-1} \leqslant (x_j + x_k) \leqslant x_{i+1},
\]

where \( \eta \) is a ratio parameter allowing us to distribute the parts of the volume resulting from the agglomeration of two particles to the closest section. This ratio is

\[
\eta = \begin{cases} 
\frac{x_{i+1} - v}{x_{i+1} - x_i}, & \text{if } x_i \leqslant v \leqslant x_{i+1}, \\
\frac{v - x_{i-1}}{x_i - x_{i-1}}, & \text{if } x_j \leqslant v \leqslant x_i.
\end{cases}
\]

This set of differential equations was solved using a fourth-order Runge–Kutta method (RK). According to the modeled particle population, an auto-adaptive time step was defined within the
RK procedure and the convergence criterion was based on the comparison between solutions obtained with RK methods of fifth and fourth order (the Cash–Karp technique). Those details are extensively discussed in the Fortran numerical recipes handbook [20].

3.3. Aerosol agglomeration: kernels

In equation (4), the most important terms that contain all the physics of the model are the agglomeration kernels $\beta_{a,b}$. These kernels are related to the probability of agglomeration of particles with characteristic volumes $x_a$ and $x_b$. Among the various mechanisms responsible for dust agglomeration, there are Brownian and/or turbulent motion, electrostatic interactions [21], etc. The determination of agglomeration kernels of carbonaceous dust present in the RF plasma is very tricky, since all the physics of agglomeration is not fully understood. In the present study, as a first step, agglomeration is solely studied taking into account Brownian motion. Due to low pressure within the experimental reactor, the mean free path of gas components is much higher than the carbon particle diameter. Therefore, the characteristic Knudsen number is very large ($Kn = \frac{2xm_f}{D_D}$); consequently dust is not driven by gaseous flow. Therefore, according to Seinfeld and Pandis [22], the corresponding kernel is

$$\beta_{1,2} = \pi (R_{p1} + R_{p2})^2 \sqrt{(c_1^2 + c_2^2)}$$

with $R_{pi}$ being the radius of the particle and $\vec{c}_i$ its mean velocity given by the Maxwell–Boltzmann distribution as

$$\vec{c}_i = \sqrt{\left(\frac{8k_B T}{\pi m_i}\right)}.$$

With this kind of kernel, agglomeration between small and large particles is clearly the dominant mechanism: small–small particles agglomeration and large–large particles agglomeration are weak due to a small collision cross-section and small velocities, respectively, as is shown in figure 3. This Brownian agglomeration kernel can be generalized to a medium with higher...
pressure taking into account gas transport through correlation involving the particle Knudsen number [22].

3.4. Light transmissivity modeling

Once the particle size distribution is known, radiative properties of the medium can be assessed. In a previous work, the light transmission measurements performed in the infrared spectra were compared to numerical simulations of our RF plasma discharge [14]. The two main features of the model, which will not be detailed here, are as follows. On the one hand, a fine description of the radiative absorption and scattering coefficients of carbonaceous dust was achieved based on Mie theory. On the other hand, light transmission through the dusty plasma was treated with a ray-tracing technique based on Monte Carlo simulation. Both parts of the model are linked by the particle size distribution, which rules optical properties of the medium and thus its transmissivity behavior.

In the present work, agglomeration simulations are used as a third model to give more reasonable input parameters to the light transmissivity modeling.

4. Simulation results

4.1. Model parameters

The first step of the numerical simulation requires one to define the discretization of the particle size range into sections. Here, particles in the range $1 \, \text{nm} < D < 2 \, \mu \text{m}$ were allocated in $N_{\text{sec}} = 200$ sections with a logarithmic scaling law. A non-uniform scaling of the particle distribution is necessary because of particle diameters ranging over three orders of magnitude. It should be noted that the smallest particles considered ($D = 1 \, \text{nm}$) correspond to tiny clusters of carbon precursors already agglomerated. They come from acetylene dissociation in the plasma discharge. The primary chemical processes leading to this agglomeration, discussed by Watanabe [23], are not taken into account in the present study. The production rate $\dot{m}$ of the smallest particles is calculated on the basis of the acetylene flow rate set during experiments. In the present case, it was assumed that $\text{C}_2\text{H}_2$ injected during experiments is converted into carbonaceous dust with $\dot{m} = 1.67 \times 10^{-6} \, \text{kg s}^{-1} (0.1 \, \text{g m}^{-3} \text{min}^{-1})$. In relation to the mass flow rate, an injection factor (IF) is defined. This last parameter is used to give a weight to the particle population in the first section $N_1$. Thus increasing IF means that more carbonaceous precursors might agglomerate, as in experiments where acetylene flow is raised.

Besides, for the upper diameter limit, it was set that particles with diameter larger than $D = 2 \, \mu \text{m}$ falling outside the plasma sheath. This limitation allows us to take into account several diameter classes, which can be expected in experiments according to the operating parameters. This is an arbitrary choice that has been validated by scanning electron microscopy measurements of deposited dust. These two extreme diameters rule the whole agglomeration process. As will be discussed hereafter, the choice of $D_{\text{min}}$ and $D_{\text{max}}$ modifies the particle population and consequently the optical properties of the dusty plasma. The following simulation results provide some examples of the impact of these choices on the evolution of the particle population.

Finally, density equations are integrated with a variable time step chosen by the fourth-order RK method. It should also be noted that the timescale is not simply the experimental one because of the agglomeration kernel assessment. It is obvious that the physics of agglomeration...
should not solely include Brownian motion but many more interactions such as electrostatic, turbulence, gravity and so on, which might affect the particle interactions and thus the global agglomeration process.

4.2. Particle agglomeration

Figure 4(a) presents a map of the particle population in each diameter class versus agglomeration time. The color scale (the z-axis) is given as the common logarithm of the population $N$, $z = \log_{10}(N)$. For this simulation the input parameters are $D_{\text{min}} = 1 \text{ nm}$ and $D_{\text{max}} = 2 \mu\text{m}$, $N_{\sec} = 200$ and the injection factor is IF = 1, which corresponds to the production rate $\dot{m} = 1.67 \times 10^{-6} \text{ kg s}^{-1}$ previously defined.

At the beginning of the numerical simulation, there are mostly small particles and the larger diameter classes are empty. Calculations point out the fact that the agglomeration process, with
the current parameters, results in oscillations of the particle density for most of the considered diameter classes. These oscillations are unvarying and a kind of periodic steady state regime, corresponding to filling/emptying processes, occurs. The agglomeration of smaller particles into larger ones is coherent with the agglomeration kernel description. The smallest particles are preferentially collected by larger ones due to the increase in collision cross-section. In order to obtain a clear representation of these fluctuations, only three representative diameters are hereafter presented. Figure 4(b) shows the normalized particle density for three classes (fine, medium and large) centered at $D = 15\,\text{nm}$, $D = 500\,\text{nm}$ and $D = 1.8\,\mu\text{m}$. The normalization was done considering the maximum density in each diameter class. From this representation it is obvious that the three densities oscillate. Furthermore, in the periodic regime, the particle density for the small diameter is not in phase with those of medium and large diameters. When the amount of small particles reaches a certain limit, these are consumed by larger ones faster than they are produced. Then the small particle density gently decreases until the moment when the medium and larger particles interact together and go beyond the $D_{\text{max}} = 2\,\mu\text{m}$ limit at which they fall. Thus, a new production cycle starts.

This type of density behavior was previously observed in the so-called ‘prey– predator’ or Lokta–Volterra models often used to describe interacting systems. For instance, binary distributions of particles with electrostatic interaction between charged particles were considered by Kim and Kim [24] and by Annaratone et al [25]. In the first study, only the first seconds of the agglomeration process were considered; in the second one, the time evolution was addressed through agglomeration rates between two classes. In our work, variations for long durations were investigated with particles that can reach diameters of some micrometers. For such dust populations, the agglomeration process cannot be solely described by two interacting classes, but necessarily by several ones.

The influence of the parameters of the numerical model was investigated. Results are reported below about the injection flow rate and the upper and lower size range limits. Figures 5(a) and (b) show the effect of the injection factor $IF$ on the evolution of two representative classes: fine ($D = 15\,\text{nm}$) and medium ($D = 500\,\text{nm}$) particles. For both particle sizes, the density behavior is the same. Whatever the value of $IF$ considered here, the density of particles in small and medium classes reaches an oscillating regime. As expected, the frequency and magnitude of the oscillations change. With larger amount of injected particles (higher IF), the density increases and the oscillation period becomes shorter. This is in agreement with experimental observations from transmission measurements of dusty plasma where the acetylene flow rate is raised [26].

During experiments, the force balance on dust strongly depends on the operating parameters [26] (pressure, delivered power, etc). As a consequence, the maximum size of particles that can be trapped in the plasma sheath varies. To test the sensitivity to the particle diameter, we modified the smallest and the largest diameter considered for injected and falling particles ($D_{\text{min}}$ and $D_{\text{max}}$), while preserving the logarithmic description of the diameter sections between 1 nm and 2 $\mu$m to allow comparison between the simulations. Accordingly, the resulting number of classes considered in the calculation varies.

In figure 6(a), the density of fine particles ($D = 15\,\text{nm}$) is addressed. This graph is subdivided into two plots due to the large variation in density according to the boundary diameter choice. In the upper plot, the lower limit is set to $D_{\text{min}} = 1\,\text{nm}$; thus there are several classes between 1 and 15 nm and the injected matter is partly distributed in each of them. In the lower plot, $D_{\text{min}} = 10\,\text{nm}$; thus, according to the volume conservation, two injected particles result
Figure 5. Influence of the injection factor on the carbon dust population versus time; $N_{\text{sec}} = 200$; diameter class, $D_{\text{min}} = 1 \text{ nm}$, $D_{\text{max}} = 2 \mu\text{m}$; injection factor: IF = 1, IF = 5 and IF = 10. (a) Fine ($D = 15 \text{ nm}$) diameter class. (b) Medium ($D = 500 \text{ nm}$) diameter class.

in one of roughly $D = 12.6 \text{ nm}$; therefore the $D = 15 \text{ nm}$ class, close to the injection class, is quickly filled and the resulting density is larger. Besides, the second noticeable point is the oscillation damping in most of the considered cases. The steady state regime observed here is quickly reached when the number of sections between the bounding diameters is reduced. Furthermore, particle numerical density is weakly varying, which means that fine particles are not fully consumed by larger ones. In consequence, the constant carbonaceous precursors injection in the first sections rapidly leads to constant density. The same remarks can be made about the medium diameter ($D = 500 \text{ nm}$) population (figure 6(b)). Nevertheless, the oscillation damping occurs later than in the previous case, since more time is necessary to fill intermediate sections.

These kinds of oscillations were experimentally observed through transmissivity and bias measurements (see the discussion in section 2.3). However, during the experiments, the
oscillation damping is not always as ‘smooth’ as in the previous calculations, because of RF discharge fluctuations that occur, e.g. dust deposition within the whole reactor, which modify the electrical parameters. This is the major disturbing effect; other erratic phenomena occurring during an experiment may also induce weak population variations.

4.3. Transmissivity of a dusty plasma in a radio frequency (RF) discharge

On the basis of the previous calculations, the spectral transmissivity of the dusty plasma was numerically assessed using ray-tracing simulation. First, the particle size distribution $N(D)$...
calculated with the sectional model and the optical properties of carbon dust (i.e. refractive and absorptive indices $n$ and $k$ [27]) were used in the frame of Mie theory to determine the absorption and scattering efficiencies ($Q_{\text{abs}}$ and $Q_{\text{sca}}$). With these data, a statistical evaluation of the radiation propagation through the plasma, taking into account the reactor geometry, was carried out. The ratio of transmitted intensity to the incident one gives the transmissivity $T_r$ of the absorbing and scattering media. Figures 7(a) and (b) show these transmissivities at several wavelengths for two particle size ranges. The injection factor is set to $\text{IF} = 10$ in order to have enough particles to affect light transmissivity in the simulations.

In both figures, it can be noted that the oscillating patterns observed on carbon dust population versus time in figures 4-6 are found again with the same periods. However, transmissivity fluctuations are smaller than population ones due to the optical properties’ progressive variation with the particle diameter. The latter properties are obtained from the whole particle distribution and averaging effects occur. A second point that should be noted
in both cases is the mean value of the transmissivity, which is about 80%. This quantity is ruled by the injection factor IF; other simulations carried with IF = 1 and 5 lead to higher transmissivities. Actually, numerical simulations provide the expected oscillating behavior of transmissivity; however, some inconsistencies remain. For example, the ordering of the transmissivities numerically and experimentally obtained differs: in figure 7(a), transmissivity at \( \lambda = 800 \text{ nm} \) is smaller than that at \( \lambda = 300 \text{ nm} \); experimentally this was never observed (figure 2(a)). When the particle size range \([D_{\text{min}}, D_{\text{max}}]\) of the carbonaceous dust is reduced, this effect becomes weaker. The calculated behavior is coherent with the experiments for long duration, when steady state is reached. Thus, the choice of the injection and falling diameters of the sectional model obviously plays a significant role in spectral transmissivity. Consequently, we modified the particle diameter boundary to observe and understand the transmissivity behavior in connection with the agglomeration model. In the following simulations, we set \( D_{\text{min}} = 1 \text{ nm} \) and \( D_{\text{max}} = 300 \text{ nm} \) for the agglomeration process. The upper diameter limit has been chosen in agreement with the mean deposited dust diameter (collected in the reactor after experiments). Once again, the reduction in the number of sections of the agglomeration model results in oscillation damping for each particle population and for the resulting transmissivity. But this time, the transmissivity greatly changes with the wavelength. The explanation of this phenomenon can be found in the variations of the extinction efficiency \( (Q_{\text{ext}} = Q_{\text{abs}} + Q_{\text{sca}}) \) plotted in figure 8(b). The extinction efficiency is related to the ability of a particle to absorb and scatter radiation. Here, it is represented as a function of particle diameter and wavelength. Briefly, high \( Q_{\text{ext}} \) values imply low transmissivity. What can be seen for the carbonaceous dust considered in our simulations is that small particles (below 100 nm) poorly affect the transmissivity, especially at high wavelengths. Their influence becomes significant only when there is a very large amount of these particles in comparison with larger ones. On the contrary, large particles (between 0.5 and 2 \( \mu \text{m} \)) strongly participate, in particular for the high wavelengths considered here. Therefore, bounding the upper diameter limit to \( D_{\text{max}} = 300 \text{ nm} \) aims at lowering the extinction at these larger wavelengths.

With these last results it can be noted that the agglomeration model combined with the Mie scattering description can be used to explain some of the main features of the experimental spectra. For example, during the first seconds of the experiments, there is a rapid decrease of dusty plasma transmissivity. This can be noted in figure 2(a) with successive minimal values of transmissivity for the three considered wavelengths. This phenomenon is well captured by the numerical simulation (see figure 8(a)) with a time delay between the first minimal transmissivities at \( \lambda = 300, 500 \) and 800 nm. Through the comparison between numerical simulations and experimental measurements, in the early stage of the agglomeration process, the production rate of the smallest carbonaceous dust might be assessed. For long durations, when the oscillating regime is observed, the period measurement associated with scanning electron microscopy analysis of dust deposits can help in retrieving the upper diameter limit \( D_{\text{max}} \). In conclusion, there are several possibilities to make use of numerical simulation in order to retrieve some of the dusty plasma key parameters. However, further work is needed to clearly identify the ones that can be appraised with good accuracy through an inversion tool.

5. Conclusion

The present study was dedicated to the understanding of dust agglomeration cycles in RF plasma discharges. In order to better understand experimental results, two numerical models...
were developed. The first one is an agglomeration model, which calculates the particle size distribution according to a discrete representation of the dust diameter spectrum into sections. The second one uses the previously computed particle size distribution to set up the optical properties of the dusty plasma using the Mie theory. Then a ray-tracing technique was applied to compute the transmissivity of a probing light through it. In the first part of the study, calculations have pointed out the significance of discretization parameters (bounding diameters) on the calculated particle populations. Different oscillating behavior of the carbonaceous dust density was observed, from sustained to damped oscillations. Besides, the rate of primary particle injection (IF) versus oscillation period and maximum density was also investigated and found to be coherent with experiments. In the second part of the paper, numerical simulations of the transmissivity measurements achieved in our RF reactor were carried out. The spectral

**Figure 8.** Transmissivity calculations versus time; $N_{sec}$ is variable according to $[D_{min}, D_{max}]$; injection factor: IF = 10; continuous line ($\lambda = 300 \text{ nm}$), dotted line ($\lambda = 500 \text{ nm}$), dash-dotted line ($\lambda = 800 \text{ nm}$); spectral extinction efficiency was obtained by the Mie theory. (a) $D_{min} = 1 \text{ nm}$, $D_{max} = 300 \text{ nm}$ (b) Extinction efficiency $Q_{ext}$. 

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behavior of the computed transmissivity clearly depends on particle size distribution variations. Several scenarios of particle injection and losses within the plasma put forward the fact that dust particles of large diameters should quickly fall to ensure a transmission behavior coherent with the experimental ones. However, calculations could still be improved by taking into account more carefully some important features such as: spatial heterogeneity of particle distribution (very large particles usually remain close to the cathode, whereas small ones can be present everywhere in the plasma sheath) and particle electrical charge (which depends on their size and on plasma parameters) in the calculation of agglomeration kernels. Some of these points are currently investigated.

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