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Nanostructured tracers for laser-based diagnostics in high-speed flows

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Abstract

The potential application of aggregates of nanoparticles for high-speed flow diagnostics is investigated. Aluminum nanoparticles around 10 nm in diameter are produced by spark discharge in argon gas. Through rapid coagulation and oxidation, aggregates of small effective density are formed. They are characterized by microscopy and their aerodynamics and optical properties are theoretically evaluated. The performance of the aggregates is experimentally investigated across an oblique shock wave in a supersonic wind tunnel of 3×3 cm² cross-section at Mach 2. Particle image velocimetry is used to quantify the time response of the aggregates. The investigations are also carried out on compact titanium agglomerates to provide a base for comparison. The results yield a relaxation time of 0.27 μ s for the nanostructured aluminum aggregates, which is an order of magnitude reduction with respect to the compact titanium nanoparticles. This work demonstrates the applicability of nanostructured aggregates for laser-based diagnostics in supersonic and hypersonic flows.

Keywords: tracer particle, PIV, shock wave relaxation time, nanostructured aggregate, spark discharge

1. Introduction

Measurement techniques such as particle image velocimetry (PIV), laser Doppler anemometry (LDA) and Doppler global velocimetry (DGV) are extensively used for non-intrusive diagnostics in flow fields. These techniques provide indirect characterization of the fluid velocity through measurement of the velocity of the suspended tracer particles. As a result, the applicability and accuracy of the tracer-based measurement techniques depend on the time response of the tracer particles, which becomes critical in extreme situations such as high-speed flows [1].

The response of a tracer particle to the velocity fluctuations in the surrounding flow field is characterized using the Stokes number, which is the ratio of the particle relaxation time to the time scale of the flow [2]. At high Stokes numbers a significant measurement error is introduced due to the slip velocity between the tracer particle and the fluid motion [3]. The slip velocity can be of *tangential* (e.g., when a particle crosses a normal shock wave) or of *transverse* type (e.g., curved streamlines around a vortex). In the latter case, the measurement errors can be felt far downstream of the region where the slip occurred due to the possible generation of particle-free regions from an integrated effect of the particle motion across the curved streamlines. The particle-free regions limit any tracer-based measurement and are commonly observed in situations such as vortices [4] and highly curved trajectories [5,14] in high-speed flows.

A sample PIV image of the streamwise wall-normal crosssection of a steady rolling vortex developed along a delta wing at free stream of Mach 2 is shown in figure 1(left). The dark area without any particle corresponds to the streamwise section of the vortex core; the particles are made to drift toward the periphery by the radial acceleration, which results in the observed absence of particles in the vortex core region.

A PIV image of flow around a scaled model of a re-entry capsule in a Mach 7.5 flow is shown in figure 1(right). No tracer particle in the area behind the capsule is detected, which is due to the inability of the particles to follow the curved path through the high-velocity and low-pressure region at the shoulder of the capsule [6]. The high curvature, high velocity and low pressure in this region contribute to the consistent



Figure 1. Visual evidence of the imposed limitation by tracer particles on PIV measurement in a vortex over a delta wing at Mach 2 [6] (left) and a capsule in hypersonic flow [14] (right).

outward ejection of tracer particles due to *transverse* slip from the region close to the object surface, resulting in a lack of tracer particles downstream in the separated flow region [7]. The *transverse* slip in this case causes the particles to move along trajectories with lower curvature. Once the particles are centrifuged away from the separated mixing layer, they will not enter the separated wake region at any downstream position.

Several types of particles and particle generation systems utilized for PIV or LDA measurements have been reported in the literature [7]. Atomization and condensation mechanisms can supply small droplets of narrow size distribution for diagnostics in gas flows of moderate temperatures [2]. Solid powders are also commonly used as tracer particles in highspeed flow and for combustion diagnostics [8]. They may be obtained from atomization of suspensions or from commercial powders, available in a diverse variety of materials and primary particle sizes. However, it is commonly reported that the agglomeration process forms particles several times larger than the primary particle sizes as a result of humidity and prolonged storage. Proper dispersion systems such as fluidized beds, cyclones and inertial impactors are required to break or filter out the coagulated particles [2, 8, 9].

In a fluidized bed, a moderate stream of gas passes through a bed of particles and suspends the smaller ones [10]. Cyclone aerosol generators apply the momentum of an impinging/swirling stream of gas to lift the smaller particles from a powder bed [11]. A rotary brush seeder breaks the coagulated particles by transporting them into a high-speed stream of gas [2]. Willeke *et al* [12] attempted to break the agglomerates by passing them through a two-phase fluidized bed of 100–200 μ m beads. Altgeld *et al* [13] have developed a mechanism to disperse the powders placed on a rotating electrode disk using the pressure waves generated by an electric spark. However, measurements under the extreme high-speed conditions mentioned above demand particles of still smaller relaxation time [6, 14].

The velocity of tracer particles across a stationary shock wave has been the most followed approach to evaluate the relaxation time of particles [15-18]. Across a shock wave, the particles follow the actual flow streamlines; however,

their velocity time history does not exhibit a discontinuity at the shock location. Instead, the particles decelerate/relax to the flow velocity downstream of the shock wave in a finite time. Lang [4] records a relaxation time of 6 μ s for oil droplets of 1.95 μ m generated by Laskin nozzles. The shock test of Haertig *et al* [17] shows a relaxation time of 2.9 μ s for TiO₂ particles of 0.4 μ m diameter. Scarano and van Oudheusden [16] measured a relaxation time of 2.4 μ s for TiO₂ particles with 270 nm nominal diameter. Ganapathisubrama *et al* [19] have recorded a relaxation time of 2.6 μ s for TiO₂ particles of 20 nm nominal size dispersed using a cyclone seeder. More recently, Ragni *et al* [18] have obtained values in the range of 0.4–3.7 μ s based on an extensive investigation on the relaxation time of different solid particles and generation/filtration procedures.

Shorter relaxation times have traditionally been aimed at by applying smaller particles. However, the minimum particle size is limited by scattered light as it declines with the power 2 of the particle diameter for micron-sized particles (Mie scattering regime) and with the power 6 for sub-micron particles (Rayleigh scattering regime) [6]. A solution to this problem can be the use of non-compact particles such as porous, hollow or chain-type, which have a low effective density.

The particles with low effective density may offer smaller relaxation time while scattering enough light to be detectable by the measurement systems. The use of non-compact particle tracers in high-speed aerodynamics has received limited attention, and to the authors' knowledge only few works can be found in the literature that refer to these particles for high-speed aerodynamics. For instance, Raffel *et al* [15] have suggested the application of porous spherical particles. This motivates the present investigation on the use of non-compact particles for particle image velocimetry in high-speed flows. Further experimental investigation is required on the performance of different varieties of non-compact particles.

The present work investigates the potential application of a new type of nanostructured aggregates with small effective density. The aggregates are generated from vaporizing metal electrodes by spark discharge. The study first includes a characterization of the aggregates using microscopy. Next,



Figure 2. Generation of fractal aggregates using spark discharge mechanics.

the aerodynamic performance of the particles is theoretically evaluated. Particle image velocimetry is used to evaluate the relaxation time of the particles across an oblique shock wave. A comparison is also proposed with commercially available TiO_2 solid tracers. In order to provide a cross-evaluation, the aerodynamic diameter of the particles is also measured using a particle sizing system. Finally, the suitability of the nanostructured aggregates as tracer particles for PIV in terms of light scattering capabilities is discussed.

2. Tracer particles properties

The nanostructured aggregates used in this work are generated by the spark discharge mechanism introduced first by Schwyn *et al* [21] and further applied and studied [20, 22, 23]. In this mechanism, which is illustrated in figure 2, two opposing cylindrical electrodes are located a few millimeters apart. Inert gas flows in the gap between the two electrodes. The electrodes are in parallel with a capacitor, which is charged by a constant current, continuously increasing the voltage between the two electrodes. When the breakdown voltage is reached, a spark discharge occurs, momentarily heating the electrodes locally to a high temperature so that the material evaporates. The vapor is subjected to rapid cooling and generates a high concentration of nanoparticles. In the present study, primary particles of aluminum, about 10 nm in diameter, are produced. They rapidly coagulate, and in the presence of oxygen form aluminum oxide aggregates of fractal-like geometry (see below). These particles are referred to as 'fractal aggregates' in this work. The nanostructured aggregates have the advantage of being easily produced and offer a broad distribution of aggregate size.

Powders of TiO₂ particles (Kemira, L830) which are typically used as tracers for high-speed flow measurement are also used in this work to provide a base for comparison. The particles have a primary crystal size of 50 nm; however, they form larger agglomerates as a result of storage and humidity. These particles are first dehydrated in an oven and then dispersed into the flow using a cyclone seeder (more details in [18]). These particles are referred to as 'compact agglomerates' in this work.

2.1. Micrographs

Images obtained using transmission electron microscopy (TEM) and scanning electron microscopy (SEM) are used to derive the geometric features of the fractal aggregates and the compact agglomerates. Two TEM images of the fractal aggregates are shown in figure 3. Pronounced necks between the primary particles indicate a rigid structure locally forming chain-like branches. Clustering of particles in an aerosol by diffusion-limited aggregation leads to structures of fractal geometry characterized by the scaling relation [24, 25]

$$N_P \propto R^{D_f}.\tag{1}$$

Here N_p is the number of primary particles that the aggregate consists of, *R* is a characteristic dimension of the aggregate, usually the radius of gyration, and D_f is the fractal dimension. For clusters formed by coagulation without reconstruction, D_f is usually smaller than 2 [25]. A fractal analysis of the aggregates observed by microscopy is consistent with $D_f <$ 2. The primary particle diameter in the agglomerates is also about $D_p = 10$ nm.

Sample SEM images of the compact agglomerates are shown in figure 4. The particles are agglomerates of primary particles of 50 nm. They are closely packed clusters and thus their effective densities may be estimated as $\rho_{\rm comp} =$ 1 g cm⁻³. It is observed that the mean diameter of the compact agglomerates can be different depending on the method of dispersion. A mean diameter of $D_{\rm comp} = 0.5 \ \mu m$ is assumed



Figure 3. TEM images of aluminum oxide fractal aggregates generated by the spark discharge mechanism. A magnified view is shown on the right-hand side.



Figure 4. SEM images of the compact agglomerates of titanium oxide. A magnified view is shown on the right-hand side.

according to the micrographs. The fractal dimension of these compact agglomerates is also estimated to be around 3.

2.2. Relaxation time

For aggregates with $D_f < 2$, the primary particles hardly shield each other in collisions with gas molecules if the gas molecules have ballistic trajectories. The no-shielding assumption exists if the mean free path of the molecules is much larger than the particle size. This assumption can be made in the present case, and the mobility of a fractal aggregate, b_{fractal} , is given by its total cross-section A_p or from the number and the diameter of the primary particles as [26]

$$b_{\text{fractal}} = \frac{U_p}{F_D} = 1.39 \frac{(kT/m_g)^{1/2}}{pA_p} = 1.39 \frac{(kT/m_g)^{1/2}}{pN_p \pi D_{\text{prim}}^2}.$$
 (2)

Here k is Boltzmann's constant, T is the temperature, p is the pressure and m_g is the mass of a gas molecule. With the no-shielding assumption, the mass of the aggregate can be estimated via N_p derived from a two-dimensional projection with

$$m_{\text{fractal}} = \frac{1}{6}\pi D_p^3 \rho_p N_p, \qquad (3)$$

where ρ_p is the density of the primary particle. Note that some higher contrast areas in figure 3 indicate that shielding occurs to a small extent so that the no-shielding assumption will somewhat underestimate the mass. The relaxation time of a particle in a gas for the laminar case (particle Reynolds number $Re_p < 1$) is

$$\tau = mb. \tag{4}$$

Combining equations (2) and (4), one obtains for an aggregate of fractal dimension smaller than 2 the following expression for the relaxation time:

$$\tau_{\rm fractal} = 0.23 \frac{(kT/m_g)^{1/2}}{p} \rho_p D_p.$$
 (5)

Thus the relaxation time is independent of the size of the aggregate in terms of the number of primary particles. This means that only the primary size has to be controlled, and there may be a distribution of aggregate sizes, all leading to the same relaxation time. The mean aggregate size, however, must be large enough to be optically detectable (see below). For primary particles of $D_p = 10$ nm with a density of $\rho_p =$

4 g cm⁻³, $p = 13\,000$ Pa and T = 150 K (corresponding to the experimental conditions described in the next section), the estimated relaxation time is $\tau_{\text{fractal}} = 0.14 \,\mu\text{s}$.

The relaxation time for a single compact spherical particle of diameter D_{comp} can be expressed as

$$\tau_{\rm comp} = 0.23 \frac{(kT/m_g)^{1/2}}{p} \rho_{\rm comp} D_{\rm comp}.$$
 (6)

Thus,

$$\frac{\tau_{\rm comp}}{\tau_{\rm fractal}} = \frac{\rho_{\rm comp} D_{\rm comp}}{\rho_{\rm prim} D_{\rm prim}}.$$
(7)

Using the estimated density of $\rho_{\text{comp}} = 1 \text{ g cm}^{-1}$ along with the diameter of $D_p = 500 \text{ nm}$ for the compact agglomerates, an approximation for the possible improvement in relaxation behavior of the fractal aggregates compared to the compact agglomerates conventionally used is obtained as

$$\frac{\tau_{\rm comp}}{\tau_{\rm fractal}} \approx 13.$$
 (8)

Equation (8) leads to a relaxation time of $\tau_{comp} = 1.8 \ \mu s$ for the titanium compact agglomerates.

Therefore, the theoretical evaluation predicts a significant improvement in the particle relaxation time upon applying the fractal aggregates. The evaluation also demonstrates that the nanostructured aggregates should not be treated using the simple concept of (apparent) density. The density of the aggregates decreases with their size but their relaxation time is independent of their size. The relaxation time basically depends on the size of the primary particles. It is concluded that the aggregates should be large enough to be optically detectable, while retaining the benefit of the small relaxation time.

2.3. Light scattering behavior

The light scattering characteristics of the fractal aggregates can be theoretically compared with an equivalent volume compact sphere in order to evaluate their detectability. The scattered signal of particles smaller than 1 μ m drops off rapidly with particle size reduction according to Rayleigh scattering [27]. The scattered signal from the fractal aggregates which have a fractal dimension less than 2 can be related to that of the primary particles using

$$I_{\rm fractal} \propto N_p D_p^6, \tag{9}$$



Figure 5. Experimental setup consisting of the supersonic wind tunnel (ST-3), the wedge and the particle injection system.

where *I* is the scattered light intensity [28]. Using volume conservation the scattered light from a fractal aggregate can be written in terms of the diameter, D_{eq} , of a volume equivalent sphere as

$$I_{\rm fractal} \propto \frac{1}{N_p} D_{\rm eq}^6.$$
 (10)

This analysis shows that the scattered light from the fractal aggregates is N_p times less than the volume-equivalent spherical particle. However, it should be kept in mind that the scattered light is also N_p times higher in comparison to the scattered light from a single primary particle. In the present study, it is experimentally shown that the fractal aggregates consisting of about 1000 primary particles 10 nm in diameter are detectable when illuminated with a pulsed laser and imaged by a Peltier-cooled non-intensified CCD.

3. Experimental setup

3.1. Flow conditions

The supersonic wind tunnel (ST-3) in the Aerodynamics Department of Delft University of Technology has been used to achieve a free stream of Mach 2. The wind tunnel has a test section of 3×3 cm² with settling chamber conditions of atmospheric pressure and at 293 K. This facility is capable of continuous operation and its relatively small test section is compatible with the low production rate of the particle generator. The total pressure and temperature are 1 atm and 293 K, which correspond to a static pressure and temperature of 13 000 Pa and 158 K in the test section. An oblique shock wave is generated in the test section using a 6° wedge as shown in figure 5.

3.2. Particle generation

A commercially available particle generator (PALAS GFG-1000 [29]) is used to generate the fractal aggregates of aluminum. A stream of argon gas is focused between the electrodes to transport the particles away from the spark region. They rapidly agglomerate downstream of the spark. The argon flow rate is in the range of $4-6 \ 1 \ min^{-1}$ during the operation.

The spark discharge frequency is set to 300 Hz. The nominal production rate of this system is about 10^8 particles s⁻¹ at a flow rate of 4 l min⁻¹ [29]. This production rate is lower than the required limit for global seeding of the entire stream of the ST-3 wind tunnel.

The setup shown in figure 5 is developed for local injection of the fractal aggregates into the flow stream. The particle generator delivers the particles/argon aerosol at 2 bar pressure to a storage vessel of 21 where the aluminum aggregates oxidize to Al_2O_3 as a result of exposure to air. The particles are discharged into the wind tunnel using an injection tube located 10 cm upstream of the wind tunnel throat. The seeded stream tube has a cross-section of approximately 1 cm² in the test section, as revealed by laser sheet visualization. The particle concentration is estimated to be higher than 1000 particles mm⁻³, confirmed by the inspection of PIV images of the fractal aggregates. The typical particle image density of 1000 particles mm⁻² is illustrated in figure 6 (left), corresponding to a light sheet thickness of 1 mm.

Measurement conditions obtained from the use of compact agglomerates are also reported in this study. The compact agglomerates are obtained from commercially available TiO₂ powders (Kemira, L830) which have a primary crystal size of 50 nm. The particles are dispersed into the flow using a cyclone seeder in which the accumulated powder on the lower wall of the cyclone is entrained by a swirling stream of air. Figure 6(right) shows a sample PIV image of the compact titanium agglomerates obtained in a larger supersonic wind tunnel ($15 \times 15 \text{ cm}^2$), where the entire flow was seeded. As a result a comparatively lower particle image density is obtained (approximately 100 particles mm⁻²). Further detail of the experiments conducted with the compact agglomerates is available in [18].

3.3. PIV measurements

A two-component PIV system has been used to conduct measurement across the generated shock wave. A Peltier-cooled 12-bit CCD camera PCO-*Sensicam* with 1376 \times 1040 pixels of 6.45 μ m pitch is used to record the light



Figure 6. Portion of the sample PIV images (750 × 450 pixels) of the fractal aggregates (left) and the compact agglomerates (right).

scattered by the tracers. A Nikon objective of 105 mm focal length is set at f-stop of 11 to provide sufficient focal depth. The magnification of the imaging system is 1.2, resulting in a field of view (FOV) of 7.3×5.5 mm² and depth of field of 1.5 mm. As is shown in the magnified view of figure 5, the edge of the FOV is aligned with the shock wave to easily decompose the measured velocity difference along the shock-normal and tangential directions. Moreover, velocity vectors at an equal distance from the shock could be averaged (along the shock-tangential direction) without the need for data interpolation. The line-of-sight of the camera is also slightly tilted (approximately 4°) with respect to the normal to the measurement plane in order to reduce optical aberration effects caused by the shock wave plane [30].

A Quanta-Ray GCR-series Nd-YAG laser (Spectra-Physics) is applied as the illumination source. This laser delivers a maximum illumination of 400 mJ/pulse per 6–7 ns pulse width at 532 nm wavelength and is operated at approximately 30% of its full power in the present experiment. The laser beam is directed toward a positive cylindrical lens of 1000 mm focal length in order to form a thin laser sheet of 1 mm thickness in the FOV of the camera. The width of the laser sheet is equal to the diameter of the laser beam which is about 7 mm. In the light scattering experiment (section 4.2), the laser beam is cut into a sheet 2 mm thick by means of a knife-edge filter, which provides a uniform light distribution within the illuminated region.

PIV recordings are obtained using the single-exposure/ double-frame mode. Synchronization of illumination and image acquisition is achieved using a LaVision programmable timing unit (PTU8) controlled by DaVis 7.2. The minimum time separation that could be reliably obtained for doubleframe recordings is 0.7 μ s. As a result, the particle images travel a distance of 57 and 28 pixels upstream and downstream of the shock wave, respectively. The images have been evaluated by spatial cross-correlation using DaVis 7.4 with interrogation windows of 32×32 pixels with 4:1 aspect ratio (elongated along the shock tangential direction) and 75% overlap. The short edge of the interrogation windows (8 pixels) is aligned normal to the shock wave to obtain higher spatial resolution. Velocity vectors are returned on a grid of 23 vectors mm^{-1} . The analysis is carried out by averaging the cross-correlation maps over the available ensemble of recordings, which enhances the reliability of the measurement under intermittent seeding conditions [31].



Figure 7. Location of the FOV relative to the shock wave and the wedge. The mean velocity vector-field on contours of normal velocity obtained using the fractal aggregates is also shown.

The velocity vector field on a background of normal velocity component obtained from 120 image pairs is shown in figure 7. The figure also shows the location of the FOV relative to the shock wave and the wedge. A visual inspection of this figure shows that the region of relaxation for the fractal aggregates is of the order of a tenth of a millimeter.

3.4. Calculation of the relaxation time

The relaxation time of the particles is quantified using an oblique shock wave test which has also been applied in the previous literature [4, 17, 32, 33]. The steep velocity gradient across the shock wave creates a velocity difference between the tracer particles and the fluid. The tracer particles decelerate slower than the fluid across the shock wave and gradually adjust to the downstream velocity. The measured particle velocity along with the theoretical solution of the fluid velocity is used to calculate the relaxation time and relative distance of the particles.

The unsteady motion of a suspended particle can be expressed in the following simplified form if only the viscous and inertia terms are considered [8]:

$$\frac{\mathrm{d}u_p}{\mathrm{d}t} = -\frac{3}{4}C_D Re \frac{\mu}{\rho d^2}(u_p - u_f) = -K(u_p - u_f), \quad (11)$$

where u_p and u_f are the velocities of the particle and fluid, respectively. In this equation, C_D , Re and ρ are the drag coefficient, Reynolds number and the density of a particle with diameter *d*, respectively. μ is the dynamic viscosity and *K* is a constant. Equation (11) is integrated over the time range



Figure 8. 2C-PIV measurement of the normal velocity of the fractal agglomerates (left) and the compact agglomerates (right) across the shock wave.

of [0 *t*] which corresponds to particle velocity in the range of $[u_{p,i} u_p]$. After rearrangement this equation results in

$$\left|\frac{u_f - u_p}{u_f - u_{p,i}}\right| = u^* = e^{-Kt}.$$
 (12)

This equation is used to define the relaxation time/distance of a particle as the time/distance required for the particle velocity difference $(u_f - u_p)$ to be reduced by a factor of 1/e after passing through the shock wave [8]. Therefore, according to this definition the relaxation time, τ , is equal to 1/*K*. Equation (12) can be written in terms of τ (instead of *K*) and further integrated over the time interval of [0 *t*] to obtain the particle location normal to the shock wave, x_n , as a function of time:

$$x_n = u_{n2}t - (u_{n1} - u_{n2})\tau(e^{-t/\tau} - 1),$$
(13)

where u_{n1} and u_{n2} represent the normal velocity of the fluid before and after the shock wave. Substituting for $t = \tau$ results in the relaxation distance, ξ , as a function of the relaxation time:

$$\xi = \tau [u_{n1} - (u_{n1} - u_{n2})e^{-1}].$$
(14)

It can be shown that both equations (13) and (14) can be approximated by a linear relationship if $M_n < 1.4$. The normal Mach number is typically lower than 1.4 for most supersonic tests conducted over the wedge of a small deflection angle. Therefore,

$$\ln(u^*) = -\frac{t}{\tau} \approx -\frac{x_n}{\xi}.$$
 (15)

The relaxation distance, ξ , can be directly estimated using equation (15) from a plot of $\ln(u^*)$ versus x_n obtained from measurement [5]. Having calculated ξ , τ is obtained from equation (14).

3.5. Aerodynamic particle sizer

Measurement of the aerodynamic diameter of the agglomerates is carried out using a commercial system in order to provide complementary data on their aerodynamic performance. The aerodynamic particle sizer (APS, Model 3321) developed by the company TSI is applied in the experiments. The APS system accelerates the particles through a nozzle and measures their time-of-flight at the exit plane of the nozzle using two overlapping laser beams [34]. The measurement range of the system covers aerodynamic diameters in the 0.523 to 20 μ m range. Particles smaller than 0.523 μ m are binned in the smallest size channel. The concentration of the particles exiting the spark generator and the cyclone is reduced by dilution to obtain a maximum concentration of 1000 particles cm⁻³ for the optimum operation of the APS system.

The aerodynamic diameter, d_a , is defined as the diameter of a sphere of unit density ($\rho_0 = 1 \text{ g cm}^{-3}$) which has the same terminal settling velocity, u_{TS} , as the particle [7]. According to the definition of particles in the Stokes regime

$$u_{\rm TS} = \frac{\rho_p d_e^2 g}{18\eta \chi} = \frac{\rho_0 d_a^2 g}{18\eta},$$
 (16)

where C_s and η denote the slip correction factor and gas viscosity, respectively. Therefore,

$$\rho_p d_e^2 = \chi \rho_0 d_a^2. \tag{17}$$

Using equations (4) and (17) and the definition of particle mobility [7], the aerodynamic diameter of a particle can be related to its relaxation time as

$$d_a^2 = \frac{18\eta}{\rho_0 C_s} \tau. \tag{18}$$

4. Results

4.1. Aerodynamic behavior

The aerodynamic behavior of particles is investigated by measuring the velocity decay of the particles across an oblique shock wave using the 2C-PIV system. The velocity of the particles normal to the shock wave, u_n , normalized using the free stream velocity before, u_{n1} , and after, u_{n2} , the shock wave is shown in figure 8 for the fractal aggregates (left) and the compact agglomerates (right). It is observed that the fractal aggregates reach the downstream velocity after traveling about 0.3 mm while the compact agglomerates require about 2 mm of traveling distance. This demonstrates that the fractal aggregates are capable of adjusting their velocity to the conditions downstream of the shock wave at a shorter distance and consequently in a shorter time.

In order to quantify the relaxation time based on the method detailed in section 6.4, $\ln(u^*)$ is plotted versus



Figure 9. Velocity decay of particles in terms of $\ln(u^*)$ for the fractal aggregates (left) and the compact agglomerates (right).



Figure 10. Aerodynamic diameter of the fractal aggregates and the compact agglomerates measured using the APS system.

 x_n in figure 9 for the investigated tracers. Using the slope of the linear fit, relaxation distances (ξ) of 77 and 500 μ m are obtained for the fractal aggregates and the compact agglomerates, respectively. The corresponding relaxation times are 0.27 and 2.0 μ s for the fractal aggregates and the compact agglomerates, respectively. This result experimentally confirms that the fractal aggregates have a shorter relaxation time and demonstrate a faster adaptation to the sharp velocity gradient imposed by the shock wave in the flow field.

The probability density function (PDF) of the aerodynamic diameter of the fractal aggregates and the compact agglomerates measured using the APS system is shown in figure 10. The smallest bin represents all the particles smaller than 0.5 μ m. It is observed that all the fractal aggregates have an aerodynamic diameter lower than 0.5 μ m. The compact agglomerates are observed to have a distribution with the median at about 1 μ m. This measurement along with equation (18) predicts the relaxation time of the fractal aggregates to be at least four times shorter than the compact agglomerates.

4.2. Light scattering behavior

The poor light scattering of sub-micron particles has often inhibited their use. An experimental investigation of the optical behavior of the fractal aggregates is necessary to evaluate their performance. The PDF of the peak intensity of particles signal recorded on the CCD sensor is shown in figure 11. This plot presents a comparison between the light scattering capabilities of the two types. It is observed in



Figure 11. PDF of the peak intensity of the scattered signal of the fractal aggregates and the compact agglomerates recorded on the CCD sensor.

figure 11 that for the fractal aggregates, the distribution of the number of counts is relatively narrow. The highest peak is observed in the lowest bin and the PDF values decrease rapidly with increasing number of counts. A broader distribution and also higher counts are observed for the compact agglomerates. The comparison demonstrates that the fractal aggregates scatter less light in comparison to the compact agglomerates. The fractal aggregates use approximately 7 bit while the compact agglomerates use 9 bit from the 12 bit range of the CCD.

4.3. Potential applications in high-speed aerodynamics

The current experiments have been conducted in a relatively small supersonic wind tunnel $(3 \times 3 \text{ cm}^2 \text{ cross-section})$ operating under steady-state conditions. A streamtube of about 1 cm² across the test section could be seeded by the fractal aggregates during a discharge of approximately 5 s as a result of 10 min accumulation of the aggregates in a 21 vessel. On a larger scale, the same system would require a longer time and vessels of larger volume for production and temporary storage of enough particles to be discharged. Based on the present hardware, the upper limit for a blow-down wind tunnel cross-section is expected to be of the order of $15 \times 15 \text{ cm}^2$ with a seeded streamtube of 10 cm² cross-section. It should, however, be kept in mind that the stability of the seeded stream requires a careful control of the injection conditions.

For larger high-speed blow-down wind tunnels, the use of fractal aggregates may become possible, reverting to particle generators based on flame or laser synthesis [35], which are reported to yield aggregates of similar geometric characteristics at significantly higher production rates.

The highest potential of the present approach is seen for applications in hypersonic conditions as produced in short duration facilities (e.g., Ludwieg tubes, shock tubes). In these circumstances, the fractal aggregates can be gradually inserted at a low mass flow rate in the pressurized vessel before the gas discharge. As a result, global seeding conditions can be achieved in the test section during the short runtime (usually of the order of milliseconds).

5. Conclusion

This paper reports a theoretical and experimental investigation on the applicability of nanostructured aggregates for highspeed flow diagnostics. The aggregates are fractal-like with low effective density and with a fractal dimension smaller than 2 and have a characteristic size of about a micrometer while being formed of primary particles of 10 nm in diameter. The aerodynamic and light scattering analysis is made in comparison with compact titanium agglomerates commonly used in supersonic flows.

The theoretical study using the concept of fractal geometries shows the potential of the investigated aggregates. The assumption of *no-shielding* for the aggregates with fractal dimension smaller than 2 predicts that they are aerodynamically similar to the individual primary particles. The scattered light from the aggregates is also theoretically estimated by the superposition of the scattered light from each primary particle, which shows an increase of the detectability of the aggregates in comparison to the individual primary particles.

The experimental investigation of the aerodynamic behavior of the particles has been conducted by measurement of their velocity across an oblique shock wave using a 2C-PIV technique. The results showed a relaxation time of 0.27 μ s for the fractal aggregates, which is approximately an order of magnitude lower than the 2.0 μ s relaxation time obtained for the compact agglomerates. The light scattering investigation also showed a smaller scattering signal by the spark discharge agglomerates, however, well in the range of the capabilities of the current PIV imaging systems.

The experimental results show a significant improvement in the relaxation time of the fractal aggregates in comparison to the compact agglomerates. This highlights the applicability of the fractal aggregates as tracers for laser-based measurement in supersonic and hypersonic flows. The ratio is smaller than that expected for the ideal theoretical case. The reason must be seen in an underestimation of the relaxation time caused by some compaction of the aggregates, leading to an increase of the fractal dimension in the vicinity of $D_f \approx 2$. This means that the no-shielding assumption is still good for an estimate but does lead to errors, as D_f may, at least in part, be larger than 2.

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