

Introduction in Nanoparticle Physics with a Focus on Combustion-derived Particles

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Abstract: A brief introduction to the behaviour of particles in gas suspension (aerosols) will be given. In particular the influence of the particle size on the interaction of the particle with the surrounding gas, which determines the motion of the particle as diffusion, sedimentation or motion of charged particles in an electrical field, is discussed. From this interaction, definitions for equivalent diameters for nonspherical particles (mobility- and aerodynamic diameters) are derived as well as definitions for the surface. The influence of coagulation on the number concentration as a function of time is shown. Furthermore, formation and properties of combustion particles are briefly discussed. Reasons leading to particle emission are given and differences between particles from different sources are discussed.

Keywords: Combustion aerosol · Equivalent diameters · Particle-gas interaction



Heinz Burtscher started his research work on aerosols at the laboratory for solid state physics of ETH with Prof. H.C. Siegmann, afterwards he became scientist at the ABB research center in Dättwil working on electrostatic precipitators for coal fired power plants. At FHNW he founded the Institute for Aerosol- and Sensor Technology. His main areas of research are characterization of small particles, in particular combustion aerosols.

1. Introduction

In the following a brief review of important properties of aerosol particles will be given. The focus is on very small particles with diameters below 500 nm, which is the size range where most particles emitted from combustion are found. Small particles with sizes in the range of the mean free path of the carrier gas and smaller behave aerodynamically very differently from larger particles (on which our intuition is usually based). As an example, mass has almost no influence on the particle dynamics. Understanding of the physics of nanoparticles therefore is essential in order to know how they can be filtered, how and where they are deposited in our respiratory system *etc.*

2. Definitions

Fig. 1 shows a typical number size distribution of atmospheric aerosols. Particles smaller than 50 nm are mostly produced by nucleation from species in the gas phase. Those in the size range from 100 nm to 1 µm result from accumulation of smaller particles. Coarse particles are often produced mechanically, for example by abrasion of tires. Particles produced by incomplete combustion mainly have diameters below 300 nm (*i.e.* they are in the Aitken or accumulation mode). Usually, particles smaller than 100 nm are called nanoparticles or ultrafine particles, whereas those in the size range from 100 nm to 2.5 µm are referred to as fine particles. However, this definition is not strictly applied.

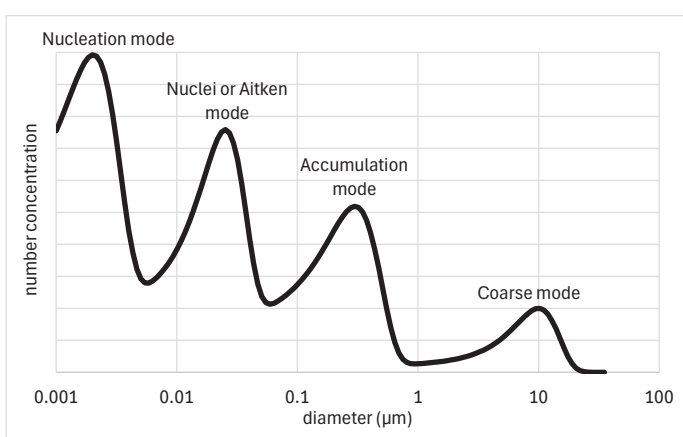


Fig. 1. Schematic representation of a typical particle size distribution of atmospheric aerosols.^[1]

3. Interaction with the Carrier Gas

In the following a brief description of some particle properties is given. Processes are discussed that are important for particle motion in the surrounding gas and on which size definitions are based.

The Knudsen number K_n is an important parameter for the interaction between particles and gas (for heat conduction, mass and momentum transfer and particle movement)

$$K_n = 2\lambda_g/d \quad (1)$$

d: Particle diameter
 λ_g : Mean free path length in the gas = mean path between collisions of the gas molecules

Two different regimes can be distinguished:

$K_n \ll 1$: hydrodynamic range, movement in viscous medium, described by the Stokes' equation.

$K_n \gg 1$: molecular range, particles treated like gas molecules.

The transition regime when K_n is of the order of one also has to be treated separately.

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3.1 Particle Movement in the Gas – Mobility Diameter and Aerodynamic Diameter

For small K_n values, (particles much larger than λ_g) the Stokes' equation provides the relationship between the force acting on a moving particle ('drag force') and the velocity of the particle relative to the gas:

$$F = 3\pi\eta dv \quad (2)$$

F : Force acting on the particle ('drag force')
 η : Viscosity
 v : Speed of movement of the particle

In this size range, the force F increases proportionally to the particle diameter d . The ratio v/F is called Stokes' mobility, b :

$$b = 1/3\pi\eta d, \quad v = b \cdot F \quad (3)$$

Mobility and driving force therefore determine the movement of a particle, for example:

- gravitational force $F_{\text{grav}} = mg \Rightarrow$ sedimentation velocity,
- Brownian motion of the molecules $\Rightarrow$ diffusion
- electrical force $F = q \times E \Rightarrow$ velocity in the electric field for charged particles (q = particle charge, E = electric field).

Stokes' law applies when $K_n \gg 1$ and velocities that are slow enough for the flow to remain laminar (almost always fulfilled for small particles).

In the molecular range ($K_n \ll 1$)

$$F = \frac{3\pi\eta d^2}{2\phi\lambda_g} v \quad (4)$$

ϕ depends on the way in which particles and gas molecules collide.^[2] In the molecular range, the force increases proportionally to d^2 . In both cases, force and velocity are proportional as long as the flow is laminar. The case of very high speeds is described by Newton's law.^[3]

In many practical cases, we find ourselves in the transition range ($K_n \approx 1$). This is usually described purely empirically by the Stokes-Cunningham correction, C_c .^[3] The Stokes mobility b is multiplied by a factor C_c , which depends on K_n .

$$C_c = 1 + 1.257K_n + 0.4K_n \exp(-1.1K_n^{-1}) \quad (5)$$

Until now, it has been assumed that the particles are spherical. Only in this case is the radius unambiguous without further definitions. For arbitrary particle shapes, the mobility is used to define the mobility diameter:

The mobility b is related to the diffusion coefficient D by the Stokes-Einstein relation^[3]

$$D = b \cdot kT \quad (6)$$

Measuring particle diffusion therefore provides the same information as the mobility. Diffusion significantly determines the behavior of very small particles (e.g. their deposition in the respiratory organs or a particle filter). Most instruments to measure the size of nanoparticles are based on this diameter. The

mobility diameter is therefore a useful definition for small particles (< 200 nm).

Mobility diameter: Diameter of a sphere that has the same mobility as the particle under consideration.

If the external force is gravity (sedimentation),

$$v = bmg \quad (7)$$

The product $b \times m$ is therefore responsible for the settling velocity of a particle.

So far, only stationary motion has been considered. For accelerated motion, inertia comes into play in addition to mobility. The more general equation of motion then becomes

$$F = \frac{v}{b} + m \frac{dv}{dt} \quad (8)$$

This linear differential equation of 1st order results in a time constant

$$\tau = mb \quad (9)$$

τ determines how fast a particle reacts to changes in force.

For example: $d = 100$ nm $\tau = 8.8 \cdot 10^{-8}$ s
 $d = 1 \mu\text{m}$ $\tau = 3.6 \cdot 10^{-6}$ s

This occurs very quickly for small particles, so the assumption of stationary motion is justified in many cases.

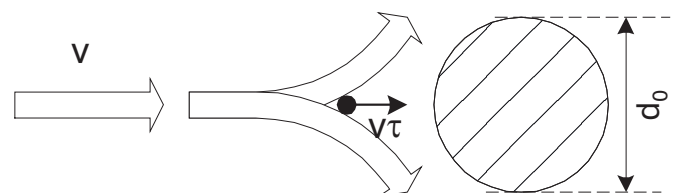


Fig. 2. Particle movement in an accelerated (deflected) gas flow, impaction: The gas flows around the obstacle, the particle flies straight ahead over a distance of approximately $v \times t = m \times b \times v$.

The same time constant τ also indicates how long it takes for a particle to reach the new gas velocity when the gas flow is accelerated. If the gas flow is deflected, particles initially continue to fly straight ahead, covering a distance of the order of $v\tau$, where v is the gas velocity. If they encounter an obstacle on this path, this is referred to as impaction.^[3] The ratio

$$s = \frac{mbv}{d_0} \quad (10)$$

of mbv and the dimension of the obstacle d_0 is called the Stokes number, s . The Stokes number determines whether a particle hits the obstacle (and is separated from the gas flow) or can follow the gas flow.

As for the sedimentation velocity, the product mb is a particle property, responsible for the impaction behavior. In addition to the mobility radius (contained in b), the mass is now also included. This is used to define the aerodynamic diameter, d_{ac} .

Aerodynamic diameter d_{ac} : diameter of a sphere of density 1 g/cm^3 , which has the same sedimentation velocity, impaction properties and mobility as the particle under consideration.

Impaction and sedimentation are dominant for larger particles. This means that the aerodynamic diameter is useful in this case, as it characterizes the particles with regard to these properties, while diffusion (determined by the mobility diameter) is less important.

The situation is completely different when a particle (or molecule) moves in a vacuum, as is the case with mass spectroscopy. As the frictional resistance then disappears ($b = \infty$), only the term

$$F = m \frac{dv}{dt} \quad (11)$$

from Eqn. (8) is relevant. This means that now only the mass is important.

3.2 Coagulation

Due to their Brownian motion particles may collide. For nanoparticles the Van der Waals potential is much higher than kT . This means that the probability that particles stick after a collision is close to unity. Agglomerates are formed, the number concentration decreases and the size increases with time. This process can be modelled by:

$$\frac{dN}{dt} = -K * N^2 \Rightarrow N(t) = \frac{N_0}{1 + N_0 K t} \quad (12)$$

where K is the size dependent coagulation coefficient, N the particle number concentration and N_0 the initial concentration.^[3] As the coagulation rate scales with N^2 , high concentrations lead to fast coagulation. Fig. 3 shows the change in concentration due to coagulation for different initial concentrations. The concentration of soot particles immediately after formation is on the order of

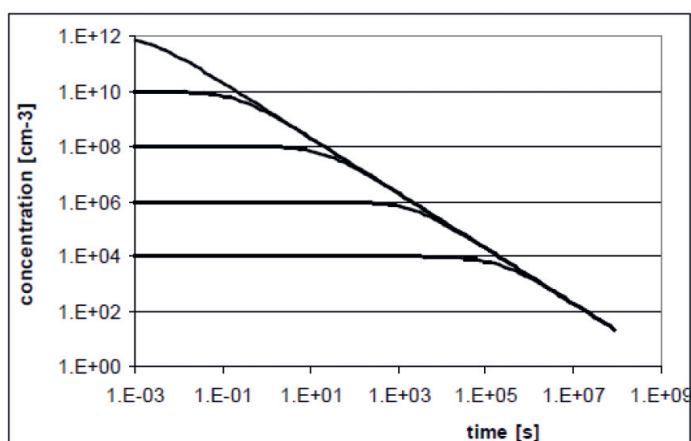


Fig. 3. Concentration as a function of time for different initial concentrations.

10^{12} cm^{-3} . Already a second later the concentration drops by more than two orders of magnitude. After sufficient time, the concentration no longer depends on the initial concentration. This simplified model assumes particles of the same size, for more information on coagulation of particles with different sizes see ref. [3]. The size distribution of aged aerosol particles in many cases is determined by the degree of agglomeration.

4. Density Mass and Surface Area

As shown above, the particle diameter for non-spherical particles must be defined by equivalent diameters. In contrast, the mass is unambiguous. The density results from the mass and the selected diameter definition. In general, the mass m of non-spherical (agglomerated) particles does not increase with d^3 , but according to

$$m \propto d^{df} \quad (13)$$

where $d_f < 3$. The density therefore decreases with increasing size. The fractal-like dimension d_f provides information about the particle structure. Various definitions for the surface are based on measurement methods. Commonly used are:

BET surface area: defined by the number of available adsorption sites.^[4]

Active surface: Defined by the adsorption cross-section.^[5,6]

Lung deposited surface area (LDSA): surface area relevant for the deposition probability.^[7,8]

The surface is important because it is responsible for many interactions with the environment, such as attachment of material from the gas phase. There is for example strong evidence that the toxicity of particles is due to such adsorbed material and the particles are the vehicle to transport them, *e.g.* in our lungs (Trojan horse effect).

5. Interaction with Light

An important class of instruments for measuring particles is based on the interaction between electromagnetic radiation, primarily light and particles.

The ratio of wavelength λ and particle radius R is important for this interaction.

I $R \gg \lambda$ geometrical optics

II $R \approx \lambda$ Mie Theory

III $R \ll \lambda$ Raleigh scattering

For visible light and fine particles, we are usually in the ranges II and III. For very small particles (range III), the intensity of the scattered light falls with R^6 and the absorption falls with R^3 . This means that in particular light scattering is very weak for nanoparticles.^[3] Information on light scattering from fractal aggregates can be found in the review written by Soerensen.^[9]

6. Combustion Particles

6.1 Formation

In a first step precursors as

- Acetylene,
- Phenylradical (C_6H_6)
- Ions ($C_3H_3^+$)
- Cyclic structures are produced by pyrolysis.

These can grow by polymerization until the supersaturation is high enough to induce nucleation. Initial nucleation particles grow by coagulation and attachment of additional material from the gas phase. Depending on temperature, availability of oxygen and time, the soot particle may be dehydrated and burn completely (Fig. 4). The yellow light in a flame is due to glowing soot particles, the resulting heat radiation cools

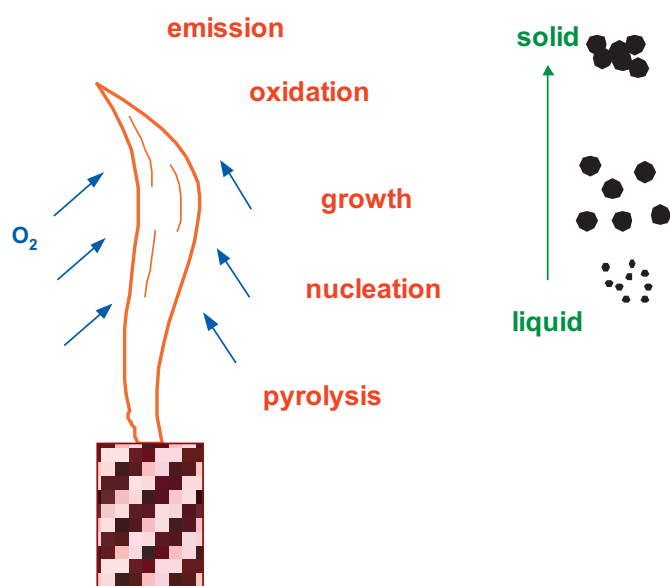


Fig. 4. Phases of particle formation during the combustion process.

the flame, the blue light of soot free (hot) flames is due to chemiluminescence.

Particles are emitted due to:

- Local or global oxygen deficiency: *e.g.* due to poor mixing of fuel and oxygen or if diffusion of oxygen to fuel droplets is too slow.
- Temperature too low: *e.g.* cooling by cylinder walls.
- Too little time for burnout: process ends abruptly when opening the outlet valve.

Here a diffusion flame is chosen to explain the principles, but they also apply for other combustion processes. The process of particle formation in combustion engines is described in Ref. [10].

6.2 Properties

The size of emitted particles is strongly determined by coagulation, *i.e.* the residence time before dilution, for example when particles exit from a tail pipe. The particles are mainly composed of elemental carbon, a variety of hydrocarbons and inorganic species like minerals and salts (ashes). The ratio strongly depends on fuel and combustion conditions. Emissions from combustion of solid fuels under good conditions, as in power plants or pellet furnaces, mainly consists of ashes (for wood mainly potassium and calcium compounds), if the combustion is rich (not enough oxygen), elemental carbon tends to dominate, if it is lean (excess air, cooling the flame) hydrocarbons are abundant.^[11]

Most particles are agglomerates and have a shell-like structure: Solid core inside (elemental carbon) on which volatile substances condense, primary particles typically are 20 nm in diameter, while agglomerates are typically 50–150 nm. Fig. 5 shows examples of diesel particles and particles from coal combustion.

Very often the particle size distributions can be approximated by a lognormal distribution. This has been shown in the work by Harris and Maricq,^[12] who developed a very useful model, allowing the estimation of properties like surface, mass *etc.* For diesel engines the distribution typically peaks in the range of 80 nm, for gasoline at 50 nm, jet engines emit very small particles in the range of 30 nm. As an example, Fig. 6 shows a size distribution of particles from wood combustion.

A significant fraction of combustion-originated particles, found in ambient air consists of secondary aerosol, *i.e.* particles formed in the atmosphere mainly by photochemical reactions from gaseous precursors.^[14,15]

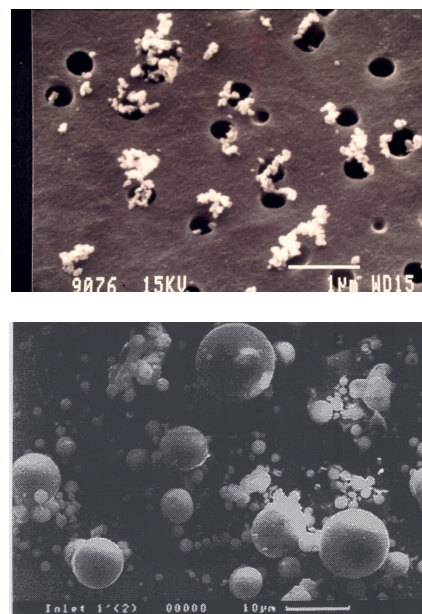


Fig. 5. Top: typical diesel particles, aggregates of primary particles. Bottom: particles from coal combustion, spherical ash particles.

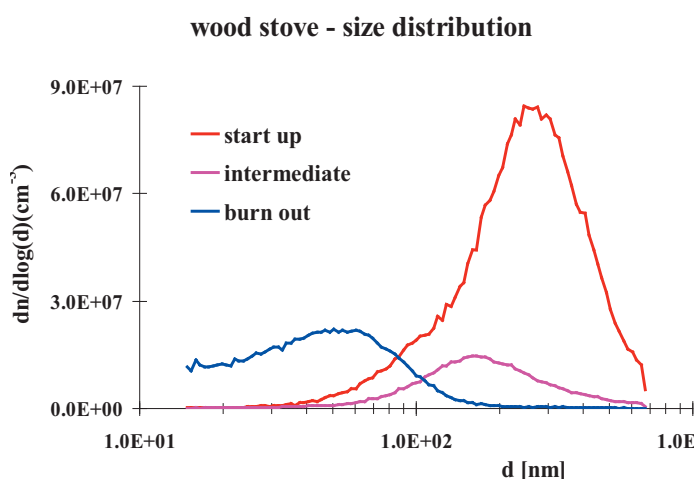


Fig. 6. Size distribution of particles from a logwood stove at different phases of the combustion.^[13]

For more information on engine emissions see, for example, the review by Kittelson.^[16] A review on particles from combustion engines is given by Überall.^[17] A comparison of size distributions from different combustion sources can be found in an article by Nussbaumer.^[18]

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