CONTROL OF PARTICULATE MATTER IN GASEOUS EMISSIONS

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Keywords: Aerosols, allergies, bulk handling, collection efficiency, cyclones, dust explosion, electrostatic precipitators (ESP), fabric filters, guided emissions, health hazard, occupational exposure, safety hazard, wet scrubbers

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Summary

In this chapter first some elementary properties of gases are reviewed. Then, the important issues of sampling and analysis are addressed. Next, the methods for characterizing particles are investigated and dust collection methods are discussed.

1. Important Properties of Gases (see, *Basic Concepts of the Gas Phase*)

1.1. Survey

Some molecular and atomic concepts are essential in understanding the physical and chemical behavior of gases.

Gases are always miscible and fill the entire space at their disposal, in contrast to the other states of aggregation, in which cohesive forces and surface tension restrain the thermal movement of molecules. **Phase changes** such as condensation and freezing, are normally accompanied by a heat effect, the **Latent Heat** of change of phase, even though they are not chemical reactions. This heat plays an important role in atmospheric cloud forming processes, buoyancy, and stability.

Most of the physical **properties** of gases, such as density ρ, viscosity μ, molecular diffusion D, heat capacity c, thermal conductivity k, and surface tension σ are intrinsic properties, as well as thermodynamic state functions. They can be derived from statistical mechanics on the basis of the kinetic theory of gases.

Temperature is related to the amount of thermal energy or heat in a system; this corresponds to the level of random motion of atoms and molecules. Temperature also governs the transfer of heat, between one system and another. Thus, one can define a scale for temperature based on the corresponding pressure and volume of the gas. In practice, such a **gas thermometer** is not very convenient, but other measuring instruments can be calibrated to this scale.

The **atmospheric pressure** is the pressure exerted by the weight of the column of air above the measuring point. Every point situated at a particular depth in a static fluid is at the same pressure.

The actual pressure, P_a, is the **absolute pressure**; the pressure difference is called **gauge pressure**. This unit is most used in technical systems, such as a steam boiler.

Finally the **kinetic theory of gases** is introduced and the concept of pressure, temperature, velocity of molecules and mean free path are revisited from this angle.

1.2. The Ideal Gas Law

The **ideal gas law** is a simple representative of an equation of state, describing the condition and physical properties of a substance as a function of generally temperature, *T* and pressure, *p*. At low densities, all gases behave ideally: on average the molecules are so far apart that their interactions can be neglected. As the density of a gas is increased, molecules come closer, interactions become tangible, and the gas starts deviating from its ideal behavior. These deviations from ideal behavior can be expressed by the virial equation of state (derived from statistical thermodynamics), the van der Waals equation (with correction factors for the volume of the gas molecules and for the intermolecular attractive forces), as well as some more recent examples of equations of state that incorporate terms taking into account the interactions between molecules and between molecules and walls.

The *Boiling Temperature* is the temperature at which the vapor pressure of a liquid equals the pressure exerted on it (usually atmospheric pressure). A substance with a higher boiling point has stronger forces of attraction, which hold the molecules together.

1.3. The Kinetic Theory of Gases

Bernoulli, a Swiss mathematician and scientist, first worked out a mathematical treatment of the kinetic theory of gases (1738), picturing gases as consisting of an enormous number of particles in very fast and chaotic motion. He realized that heat makes gas particles move faster and assumed that gas pressure is caused by the impact of particles on the walls of a container.

Clausius, Maxwell and Boltzmann introduced sophisticated mathematics into physics. Maxwell used probability to produce a distribution curve for the velocities of gas molecules and also provided a mathematical basis for Avogadro's theory. Gas particles are tiny compared to their space, in constant motion, and their interactions are very brief. The motion of particles is not only related to pressure, volume, and temperature, but also to properties such as heat conductivity, viscosity (resistance to flow), and

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diffusion

Thus, molecules carry the mechanical quantities of energy, momentum, and mass. These are called transport properties, and their rate of transport is dominated by collisions between molecules, which are described by the laws of mechanics and force their trajectories into tortuous shapes.

1.4. Thermodynamics

Thermodynamics provides a framework to the study of any process, whether physical, thermal, or chemical. The expansion of rising hot air in the atmosphere and the condensation of water vapor to cloud droplets are important processes in the dispersion of air pollution and the self-cleaning effects in the atmosphere. The partition of semi-volatile pollutants, such as PAH and PCDD/F, between atmospheric air, aerosols and dust, and the exchanges with water and soil are difficult to explain without a sound thermodynamic basis. Thermodynamics also determines the pathways numerous pollutants follow, eventually conveyed from ambient air to the blood circulating in the lungs, or reporting to the human food chain.

Thermodynamics delves into a wide range of topics including:

- Equations of state, i.e. *PVT* relationships and gas laws (both ideal and non ideal);
- Phase equilibria, e.g. those referring to water/steam or to moist air (Mollier diagrams);
- Energy balances, heats and equilibrium of chemical reactions, and combustion reactions;
- The efficiency of power plant, engines and turbines, pumps and refrigerators.

In general, thermodynamics deals with the inter conversion of various kinds of energy including heat and the corresponding changes in physical properties and phases. It is closely related to statistical mechanics from which many thermodynamic relationships can be derived.

The **law of conservation of energy** states that energy can neither be created nor destroyed; it can merely be changed from one form of energy to another. Energy often ends up as heat, which is thermal energy of atoms and molecules. The **principle of conservation of mechanical energy** states that the total mechanical energy in a system also remains constant, as long as the only forces acting are conservative forces, i.e. excluding friction losses.

1.5. Thermochemistry

Thermochemistry can be used to predict the extent to which chemical reactions proceed as well as the heat of reaction evolving as a function of operating conditions. Thermochemistry also describes the evolution of three major state functions in a reaction:

- **Entropy** (*S*),

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- **Gibb's Free Energy** (*G*), and
- Heat.

Entropy is a measure of disorder (and constantly increasing in the universe), *Heat* is a measure of the energy of the reaction and *Gibb's Free Energy* is a measure of work that must go into a reaction to make it occur (when *G* is negative) or work that a reaction can do (when positive).

1.6. Flow

In technical processes the flow regime may be either viscous (also termed streamline or laminar flow), or turbulent (also termed “eddy” flow). A viscous fluid flow regime is only encountered at low fluid velocity *v*, as in arteries or lubrication; at a given point the fluid maintains a predictable velocity and direction and its motion can be represented by streamlines. Turbulent flow is encountered at higher values of the fluid velocity; fluid velocity and direction at a given point fluctuate erratically. A flow can be moved by:

- Gravity forces, i.e. from a higher to a lower position,
- Pressure, supplied using pumps, compressors, blowers, or fans, or
- Momentum transfer (as e.g. in ejectors).

Flow patterns can be computed on the basis of three equations:

- **Equation of continuity**, a mathematical formulation of a mass balance over a stream tube, or over an infinitesimal element of volume.
- **Equation of motion**, a balance of momentum over a stream tube, or over an infinitesimal volume.
- **Equation of energy**, a balance of mechanical energy under its various forms, over a stream tube, or over an infinitesimal volume.

Steady-state conditions are conditions, invariable in time.

Advection is a horizontal flow of pollutants, driven by the wind. Vertical mixing is mainly determined in the atmosphere by the vertical temperature gradient, which determines atmospheric stability.

1.7. Transport Phenomena

Transport Phenomena describe

- the tendency of temperature, concentrations, and fluid velocity to strive towards uniform values in any physical system, as well as
- the rates corresponding with this evolution.

A similar form of mathematical equation controls these three processes, commonly termed: heat transfer, diffusion or mixing, and internal fluid friction. This explains why a unified approach is proposed for their study.

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The flow of heat, mass or momentum (and electrical charge) follow empirically derived laws of identical form, similar also to Ohm's Law for electric current. A flux is defined as a flow rate per unit cross-section. Resistance values to flow may either be additive, when presented in series, or else their inverse values (conductance values) are additive, when resistances show up in parallel.

The flow rate is a ratio of a driving force ΔV to a resistance R:

Flow Rate = Flux (Surface of the conducting cross-section)
= (Driving Force)/Resistance

This generic diffusion equation is time-dependent, i.e. it also applies to non-steady-state situations. The Resistance against flow is proportional to the length, L of the conductor times a resistance coefficient ρ, and divided by the cross-section A of the stream tube:

$$R = L\,.\,\rho/A \qquad (1)$$

The inverse of the *Resistivity*, ρ is a physical property, equal to the *Conductivity*, which is named diffusivity, D in mass transfer, thermal conductivity, k in heat transfer, viscosity, μ in momentum transfer, or electrical conductivity, ε in Ohm's Law. The value of these coefficients can all be derived from the kinetic theory of gases.

Depending on the physical situation, these empirical Laws bear different names:

- Matter diffusion is governed by Fick's Law,
- Conduction, or thermal diffusion, by Fourier's Law,
- Diffusion of momentum by Newton's Law, and
- Diffusion of electrons in an electrical field by Ohm's Law.

1.8. Conclusions

Gases can be described by the ideal gas law, or by state equations that account for the volume of molecules proper, and their mutual interactions, or with the wall.

Important properties are the transfer coefficients viscosity, heat conductivity, as well as diffusion.

Moreover, the partition between phases is a cardinal factor in the effects and spreading of pollution.

2. Air and Emission Sampling and Analysis (see, *Emission Sampling and Analysis*)

2.1. Effluent Monitoring

Gaseous effluents, such as flue gases and off-gases, must be monitored carefully in order to control:

- Plant operation,

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- Emission of pollutants, and
- sometimes both.

Either oxygen or carbon dioxide is monitored, in order to verify the excess of combustion air. This factor determines the thermal efficiency of the boiler plant. The quality of combustion (addressed in detail in *Pollution Control through Efficient Combustion Technology*) can be derived from few parameters, such as CO, TOC or sooting (Ringelmann or Baccharach value).

Pollutants proper may be measured on the basis of a wide range of general emission parameters. Frequently covered are the values for dust, heavy metals, CO, SO_2, NO_x, C_xH_y, TOC, and HCl (for incinerators). Rarely measured are SO_3, Cl_2 and N_2O. Specific compounds are either process or raw materials related, e.g. fluorides from enamels, phosphoric acid or aluminum production.

The nature and limit values of the parameters to be monitored and the method and frequency of their determination are given in general codes and also in specific conditions, stipulated in operating permits. Detailed procedures vary from country to country, although E.U.-Directives and Federal Codes may decide on minimum requirements and also on measures to be taken (notification, even halting plant operation) in case limit values are being exceeded for a specified time period.

Major Organic compounds of interest are:

- Total Organic Carbon, a sum parameter of organics in a gas stream, as monitored by a Flame Ionization Detector (FID), without further identification of the compounds involved. In TOC hydrocarbons have a higher weighing factor than compounds containing oxygen, nitrogen, sulfur, halogens, etc.
- Specific volatile organic compounds (VOCs), such as solvents, some of which are chlorinated, benzene, toluene, aromatics, paraffins, olefins (the scientific or International Union of Applied Chemistry, IUPAC names are: arenes, alkanes, alkenes).
- Polycyclic Aromatic Hydrocarbons (PAH), typically the sixteen ones identified by the US EPA, or the Borneff PAHs.
- Polychlorinated biphenyls (PCB), a group of 209 different biphenyls, substituted from once to ten times by chlorine.
- PCDD/PCDF, i.e. Polychlorinated dibenzo-p-dioxins (PCDD) and dibenzofurans (PCDF), together generally abridged to “Dioxins”, thus referring to a group of 75 different dibenzo-p-dioxins and 135 different dibenzofurans, substituted from once to eight times by chlorine. Routinely, only tetra- to octachlorinated congeners are considered, with emphasis on those with a planar structure, and characterized by a 2,3,7,8-substitution pattern as well as a toxicity equivalent factor. The NATO factors (NATO-CCMS I-TEQ) are used most. The WHO factors also take the TEQ-values of PCBs into account.

2.2. Immission Values

Generally, different procedures are required to measure immission values. The latter are

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typically up to five or six orders of magnitude lower than the corresponding emission values.

Immission values are normally not related to the emissions of a single plant, but much more to the prevailing atmospheric conditions, such as stability and wind velocity that determine the vertical mixing and advection of pollutants in the atmosphere.

Still, authorities may decide on general measures, such as informing air pollution sensitive groups in the population, limiting traffic, forced switching to a cleaner fuel, etc.

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Biographical Sketch

Alfons Buekens was born in Aalst, Belgium; he obtained his M.Sc. (1964) and his Ph.D (1967) at Ghent University (RUG) and received the K.V.I.V.-Award (1965), the Robert De Keyser Award (Belgian Shell Co., 1968), the Körber Foundation Award (1988) and the Coca Cola Foundation Award (1989). Dr. Buekens was full professor at the Vrije Universiteit Brussel (VUB), since 2002 emeritus. He lectured in Ankara, Cochabamba, Delft, Essen, Sofia, Surabaya, and was in 2002 and 2003 Invited Professor at the Tohoku University of Sendai.

Since 1976 he acted as an Environmental Consultant for the European Union, for UNIDO and WHO and as an Advisor to Forschungszentrum Karlsruhe, T.N.O. and VITO. For 25 years, he advised the major industrial Belgian Bank and conducted more than 600 audits of enterprise.

Main activities are in thermal and catalytic processes, waste management, and flue gas cleaning, with emphasis on heavy metals, dioxins, and other semi-volatiles. He coordinated diverse national and international research projects (Acronyms Cycleplast, Upcycle, and Minidip). Dr. Buekens is author of one book, edited several books and a Technical Encyclopedia and authored more than 90 scientific publications in refereed journals and more than 150 presentations at international congresses. He is a member of Editorial Boards for different journals and book series.

He played a role in the foundation of the Flemish Waste Management Authority O.V.A.M., of a hazardous waste enterprise INDAVER, and the Environmental Protection Agency B.I.M./I.B.G.E. He was principal ministerial advisor in Brussels for matters regarding Environment, Housing, and Classified Enterprise (1989). Since 1970 he has been a Member of the Board of the Belgian Consumer Association and of Conseur, grouping more than a million members in Belgium, Italy, Portugal, and Spain.

He is licensed expert for conducting Environmental Impact Assessments (Air, Water, Soil) and Safety Studies regarding large accidents (Seveso Directive).

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