TRANSPORT PROPERTIES OF FLUIDS

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Contents

Summary

The transport properties (viscosity, thermal conductivity and diffusion coefficient) of liquids and gases (fluids) are important for the most efficient engineering design of many processes in the oil, chemical and biotechnological industries. They characterize the response of a fluid to changes in its temperature, speed of flow or composition. The Chapter sets out the various ways that have been employed over more than a century to satisfy the demands of the process industries for more and more accurate information on these properties. We emphasize the scale of the task to satisfy that demand over a wide range of temperatures and pressures for an increasing number of pure components and their mixtures as the products of industry have become ever more sophisticated. Thus, we consider the major experimental techniques that have been successfully employed for direct measurement of these properties and the supporting theoretical basis that has enabled the robust extension of a finite set of measurements to a wider series of materials and circumstances. Neither the techniques nor the theory are treated in depth and rigorously, instead we exemplify the techniques and their application while giving

extensive references from which an interested reader can reach into the subject to whatever depth is required.

We also point out that because these properties reflect the underlying interactions of the molecules that make up the fluid, their study has also led to scientific insight into the forces that act between simple molecules and are responsible for the behavior of all macroscopic samples of matter.

1. Introduction

Transport processes are the processes whereby mass, energy, or momentum are transported from one region of a material to another under the influence of composition, temperature, or velocity gradients. If a sample of a material in which the chemical composition, the temperature, or the velocity vary from point to point is isolated from its surroundings, the transport processes act so as eventually to render these quantities uniform throughout the material. The non-uniform state required to generate these transport processes causes them to be known also as non-equilibrium processes. For any given gradient of a quantity such as temperature, the equilibration after isolation from the surroundings occurs through transport processes and its rate depends upon the properties of the material known as transport properties. For a large class of materials, the laws that govern the transport processes are quite simple. There are a rather large number of transport properties in principle, but three have, by far, the greatest practical and scientific importance. They are the diffusion coefficient, the viscosity and the thermal conductivity and are associated with the transport of mass, momentum and energy respectively.

1.2. Importance of Transport Properties

In practical engineering terms, the speed of the transport processes described above, and thus the magnitude of the transport properties, describes how quickly, for example it is possible to heat a fluid flowing through a heated pipe. As Figure 1 shows, during the passage of an initially cold liquid over a plate heated to a constant temperature along its length, the fluid has heat transferred to it by (energy) conduction down the temperature gradient perpendicular to the wall.

It is clear that the further along the plate the fluid flows it becomes hotter at a given perpendicular distance from the plate. Thus, if we said we wanted to heat the fluid to a particular temperature, this would determine how long the plate had to be for a given plate temperature. If the properties of the fluid are altered (specifically the thermal conductivity in this case) the length of the plate required would itself be different.

This is an example of a simple heat exchanger which is a common piece of equipment in the chemical process industry and in many domestic hot-water boilers. More sophisticated versions of this concept are used in practice but the fact remains that to achieve a particular state of the fluid will require a size of heat exchanger that depends upon the fluid transport properties. It is for this reason that the transport properties of liquids and gases, among other quantities, are required for the proper and efficient design of all equipment from a domestic water heater to a methanol synthesis reactor in a chemical plant.

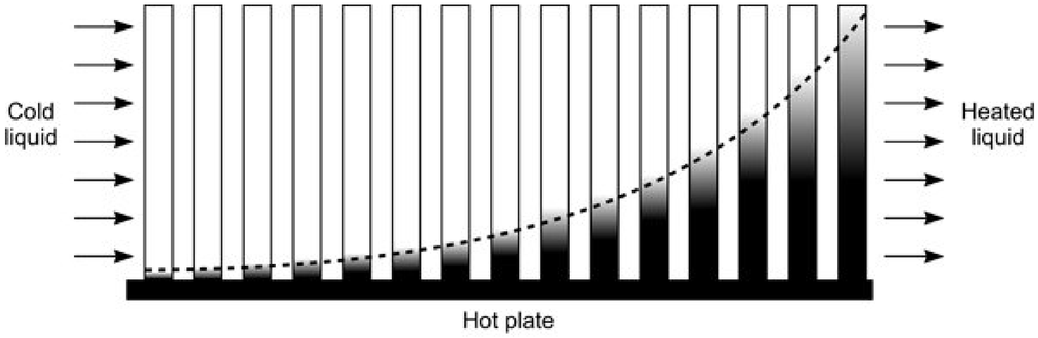

Figure 1. Cold liquid heated by passage over a hot plate

1.2.1. International Bodies/Committees and Reference Values

In engineering practice one always wishes to design a system to perform the task needed with something that is no bigger than absolutely necessary. This condition secures the greatest efficiency and therefore the most sustainable system. It follows that in a real case, such as the methanol synthesis reactor sketched in Figure 2, we must know the properties of the fluid streams with high accuracy if we are to make sure that the equipment is neither too small nor too large.

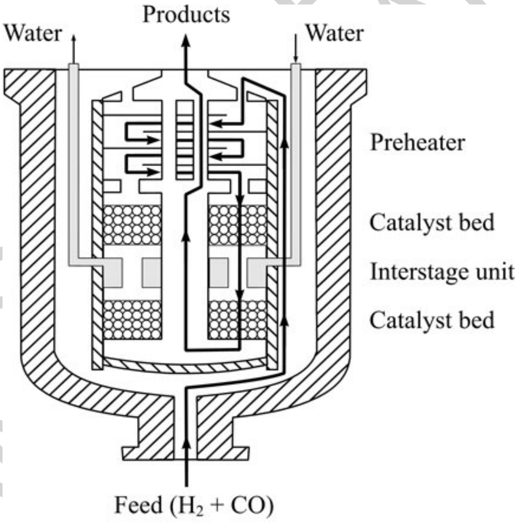

Figure 2. Methanol synthesis reactor

In such a reactor we wish to combine hydrogen and carbon monoxide over a catalyst at high temperature to produce methanol having heated them from ambient temperature. As the sketch shows, it follows that we must know the transport properties of all the streams of various compositions of at least three components over a wide range of temperature and pressure.

This suggests that a knowledge of such properties must be gained by measurement, theoretical calculation or guessing. It is far too important and too risky to use guesses so that there has been an enormous effort over a century now to seek to measure the properties of materials for these purposes (Wakeham *et al.* 1991). However, the sheer scale of the problem posed by the number of chemicals in regular use, the conditions of application and the variability of composition in mixtures, means that the industrial needs can never be met by measurement alone and there has had to be an equally large complementary effort to produce a theoretical description of fluid (gas and liquid) properties. We shall say something about both of these efforts and their outcome below.

For a limited number of materials that are in exceedingly common use such as water, steam, methane, air, hydrogen for example, it has been deemed worthwhile over many decades to seek international agreement on the properties of materials over ranges of conditions. This agreement facilitates international trade because the property values used in the design of any plant are a part of the design and this may be traded internationally. It is obviously sensible for those property values to be agreed independently of a particular problem and several international bodies have been created in the last century to provide such agreed values. In the area of transport properties, this refers to supranational bodies that propose such standards. Such bodies include the following:

- International Association for the Properties of Water and Steam (IAPWS)
- International Association for Chemical Thermodynamics (IACT)
- International Association for Transport Properties (IATP)
- International Union of Pure and Applied Chemistry (IUPAC)

International accepted values, or standard, or reference values can be found in reference journals or textbooks concerned with reference data, for example, the Journal of Physical and Chemical Reference Data to name but one. The work performed by the National Institute of Standards and Technology (NIST) on reference materials should also be mentioned.

2. Viscosity

2.1. Introduction

The viscosity of a material is a measure of the extent of irreversible dissipation of energy caused when a fluid is forced to flow through any physical configuration that involves boundaries that are not moving at the same velocity. The phenomenon of viscosity reflects the underlying microscopic interactions between the molecules of the material and is relevant only to the liquid and gaseous states of matter. In very many practical circumstances the viscosity does not depend upon the features of the flow field and then the fluid is said to be Newtonian for a reason explained below.

2.1.1. Definitions

For fluids that obey Newton’s Law, the viscosity, μ, is defined by the relationship

$$\sigma_{ij} = \mu \frac{du}{dx} . \qquad (1)$$

In this equation, σ_{ij} is the instantaneous stress at a point in the material and (du/dx) is the gradient of fluid velocity at the same point in the material. The viscosity depends on the thermodynamic state of the fluid and it is usually specified by the pairs of variables (T, P) or (T, ρ) for a pure fluid, to which must be added a composition dependence in the case of mixtures.

2.2. Measuring Techniques

Although Eq. (1) incorporates the viscosity, it does not immediately suggest means of measurement (using a device called a viscometer) because it is impossible to measure local shear stresses. It should further be pointed out, that almost all practical viscometers have a shear stress that is not constant through the fluid.

Thus, the measured shear stress is usually measured at one location but the rate of strain is not known at the same location. Methods of measurement of the viscosity must therefore be based on the determination of some integral effect of the stresses amenable to precise measurement in a known flow field.

In the first three types of viscometers considered, a solid body is forced to perform oscillations while immersed in the fluid. The characteristics of the oscillation are determined by the viscosity of the fluid.

In the two other types of viscometers non-oscillatory flow is employed, in one case by the fluid flowing through a capillary tube, and in the other a body of revolution (with cylindrical or spherical symmetry) falls under gravity through the fluid. Each has its merits and disadvantages, which are discussed.

2.2.1. Oscillating-Body Viscometers

Oscillating-body viscometers consist of an axially symmetric body suspended from a torsion wire so that the body performs oscillations in the fluid about its axis of symmetry. The oscillator can be a disc (oscillating freely or between fixed plates - see Figure 3a), a sphere (see Figure 3b) or a cylinder (cup - see Figure 3c). The fluid usually surrounds the oscillator but, in the case of the sphere or the cup, it can be inside them. The suspension wire is elastic and the system is gently rotated to start a motion.

The fluid exerts a viscous drag on the oscillator causing the frequency of the oscillation, ω, and the damping, Δ, of the resulting simple harmonic motion, to be different from those in vacuum, ω_0 and Δ_0. The magnitude of the change depends on the viscosity and on the density of the fluid in addition to the physical characteristics of the oscillator. Measurements of ω, ω_0, Δ, and Δ_0 can give the viscosity of the fluid with very small uncertainty, if the density is known from an independent determination.

The only measurements necessary in this technique are those of mass, length and time, in addition to those of the pressure and temperature, all of which can be made with a very high resolution. It is relatively straightforward to make the induced perturbation small, so that a near-equilibrium state is maintained.

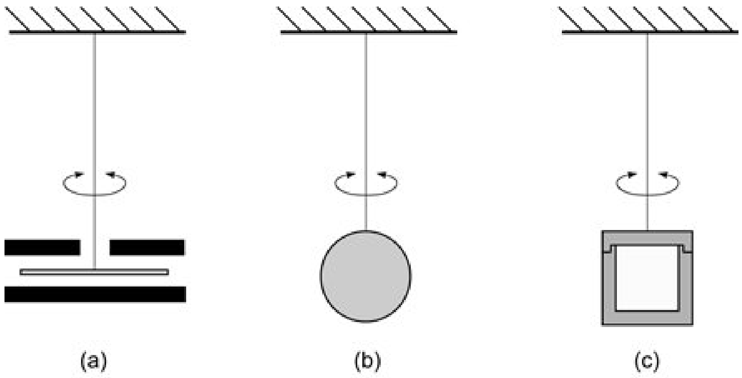

Figure 3. Types of oscillating-body viscometers

The characteristic equation for the torsional motion of any oscillating body viscometer can be expressed (Newell 1959, Wakeham *et al.* 1991) as

$$\left(s+\Delta_0\right)^2+1+D(s)=0\,. \tag{2}$$

Here, $D(s)$ is the torque on the body, calculated from solutions for the fluid flow close to the oscillating body. It is therefore the torque $D(s)$ that is characteristic of the particular type of oscillating viscometer employed.

2.2.1.1. Oscillating-Disk Viscometers

In 1959, Kestin and Leidenfrost developed one of the first very successful oscillating-disk viscometers to measure the viscosity of gases near room temperature, in the pressure range 0.1 to 60 MPa.

The basic design of such a viscometer, (see Figure 3a) consists of a disk of radius R, thickness d and moment of inertia I, oscillating between two parallel fixed plates at distances b_1 and b_2 from its surfaces. The configuration between two fixed plates has generally been preferred to a free disk, because the presence of the two parallel fixed plates tends to increase the viscous drag on the disk, and hence produce a decrement that is easier to measure with high accuracy. The plates also reduce the likelihood of influence from spurious free convective flows within the bulk of the fluid.

In principle, under a limited set of conditions, this instrument permits measurements to be made for which nothing other than the geometry of the disk and the frequency and decrement need to be measured. In particular the viscosity of no fluid is required for calibration. More generally measurements in which a calibration has been performed require less effort to achieve a comparable accuracy (Wakeham *et al.* 1991).

There has been a large series of oscillating-disk viscometers. As already mentioned, Kestin and Leidenfrost in 1959 developed the first instrument to measure the viscosity of gases near room temperature, in the pressure range 0.1 to 60 MPa, with an uncertainty of about 0.1%. One of the most successful such instruments which is still in operation today is the quartz oscillating-disk viscometer designed by Vogel in 1972 for measurements on gases and vapors up to 650 K, with an uncertainty of 0.1 to 0.3%.

2.2.1.2. Oscillating-Cylinder Viscometers

The thick disk or cylinder configuration consists of a cylindrical body oscillating in an essentially infinite fluid. Torklep and Oye in 1979 have described an oscillating-cylinder viscometer (see Figure 3c) designed for absolute measurements of low viscosity fluids up to temperatures of 1200 °C. The uncertainty quoted was 0.1% for water, where special efforts were made to achieve high accuracy, rising to 1% for routine measurements employing time-saving procedures.

2.2.1.3. Oscillating-Cup Viscometers

An oscillating-cup viscometer consists of an axially symmetric cylinder (cup - see Figure 3c) of inner radius R and height H, with the fluid contained inside. Beckwith and Newell in 1957 obtained the expression for $D(s)$ for the cases of a filled or partially filled cup (Beckwith and Newell 1957). There are many successful oscillating-cup viscometers. Grouvel *et al.* in 1977 measured the viscosity of mercury from 20 to 260 °C with an uncertainty of 1.5% (Grouvel *et al.* 1977), while Knapstad *et al.* in 1989 measured the viscosity of many hydrocarbons up to 150 °C with an uncertainty of 0.5%.

2.2.1.4. Oscillating-Sphere Viscometers

In this type of viscometer, the fluid either fills a suspended, spherical shell or surrounds a suspended, solid sphere (see Figure 3b). Accurate expressions exist for the viscous torque in either case, but the viscometer has not been as successful as have other types. This is partly because of the difficulty of manufacturing a sufficiently precise spherical shell, but also because the device offers little advantage over the simpler cylindrical viscometer.

2.2.2. Vibrating Viscometers

The essential characteristic of the oscillatory viscometers discussed in the previous section was the observation of the effect of the viscosity of the fluid on the damping of the free, torsional motion of a rigid, solid body either immersed within it or surrounding it. In the case of the vibrating-wire viscometers, the oscillations involve periodic distortions of the solid body itself, which is in contact with the fluid whose viscosity is under investigation. The main advantages of vibrating viscometers are that the instruments are mechanically simpler than those of oscillatory bodies, and that the volume of fluid required for their use is much smaller, making operation at extremes of higher pressures and temperature easier.

2.2.2.1. Vibrating-Wire Viscometer

The vibrating-wire viscometer is a particularly simple form of vibrating viscometer, which has successfully been employed over a wide range of conditions. A thin, circular-section wire of radius R, density ρ_s, subject to a tension, is constrained to be stationary at either end (see Figure 4a). It is surrounded by an infinite volume of the fluid of interest and performs oscillations transverse to its axis in a single plane containing the axis. If the oscillation is initiated by an initial deflection at time $t = 0$ and subsequently allowed to decay, then the displacement of any point on the wire, ξ, ultimately conforms to a damped, simple harmonic motion (Retsina *et al.* 1987)

$$\xi = Ae^{-\Delta\omega t}\sin(\omega t + \varphi)\,, \tag{3}$$

in which ω is the angular frequency and Δ the logarithmic decrement. In essence the system resembles the plucking of a guitar string. The motion of the wire is readily detected and measured electromagnetically so that the decaying signal is easily recorded. Under appropriate conditions (Retsina *et al.* 1987), the decrement and frequency of the motion can be related to the viscosity μ, and density ρ, of the fluid. The same device can also be operated as a driven resonator and the characteristics of the resonance related to the same properties of the fluid.

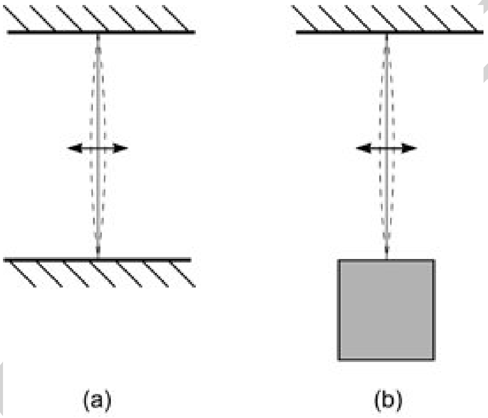

Figure 4. Vibrating-wire viscometers

In favorable cases it is possible to perform absolute measurements of the viscosity of fluids by either version of the technique although a calibration is easily performed which allows simpler operation.

With a slightly modified design, in which the tension in the wire is generated by means of a mass suspended on the wire (see Figure 4b), it is also possible to measure the density of the fluid simultaneously. Only one correction to the working equations given above is significant (>0.01%) and that arises from the fact that it is necessary to enclose the fluid within a solid wall. A correction for this effect has been derived (Retsina *et al.* 1987) and it is rather straightforward to allow for the presence of the boundary.

The precision in the viscosity measurements for liquids is about ±0.1%. For measurements in gases, the uncertainty is about ±1% when operated in an absolute way, and better than ±0.5% when operated in a relative manner. Vibrating-wire viscometers have successfully been operated over a wide range of conditions (Assael *et al.* 1991, Oliveira 1991, Correia Da Mata *et al.* 2001, Wilhelm and Vogel 2001).

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Biographical Sketches

Konstantinos D. Antoniadis studied at the Chemical Engineering Department of the Aristotle University of Thessaloniki, where he received his diploma in 2004 and his Ph.D., on the Measurement of Thermal Conductivity of Composite Solid Materials, in 2011. He is author and co-author of 11 publications in referred journal and in several proceedings of international scientific conferences. His research activities are mainly focused on the area of the measurement of thermal conductivity of liquids, nanofluids and solids. He is currently a researcher of the Laboratory of Thermophysical Properties & Environmental Processes of the Aristotle University of Thessaloniki

Marc J. Assael received his PhD from Imperial College in 1980. In 1982 he was elected Lecturer in Heat Transfer in the Faculty of Chemical Engineering at the Aristotle University of Thessaloniki, where he founded the Thermophysical Properties Laboratory. In 2001 he was elected Professor of Thermophysical Properties at the same Faculty. During the years 1991-1994 he served as the Vice-Chairman of the Faculty, during 1995-1997 as the Chairman of the Faculty, and from 2009-today again as the Vice-Chairman of the Faculty of Chemical Engineering.
In 1998, Marc Assael was TEPCO Chair Visiting Professor in Keio University, Tokyo, Japan, while during the years 2007-2010 he also held the position of Adjunct Professor in Jiaotong University, Xi'an, China. He has published more than 230 papers in international journals and conference proceedings, 20 chapters in books and 7 books. In 1996, his book Thermophysical Properties of Fluids: An Introduction to their Prediction (coauthored by J.P.M. Martin & Th.F. Tsolakis) was published by Imperial College Press, while in 2010, his latest book, Fires, Explosions and Toxic Gas Dispersions: Effects Calculation and Risk

Analysis (coauthored by K. Kakosimos), was published by CRC Press. He is acting as a referee for most journals in the area of thermophysical properties, while he is also a member of the Editorial Board of the scientific journals

- International Journal of Thermophysics (Kluwer Academic Publishers, Plenum Press)
- High Temperatures High Pressures (Pion, UK)
- IChemE Transactions Part D: Education for Chemical Engineers (IChemE, UK)
- International Review of Chemical Engineering - Rapid Communications (Praise Worthy Prize, IT)

Marc Assael is a national delegate in many committees in the European Union, in the European Federation of Chemical Engineering as well as in many international scientific organisations.

Professor Sir William Wakeham retired as Vice-Chancellor of the University of Southampton in September 2009 after 8 years in that position. He received his PhD in Physics at Exeter University in 1969. In 1971, after a postdoctoral period in USA at Brown University, USA he took up a lectureship in the Chemical Engineering Department at Imperial College London and became a professor in 1983 and Head of Department in 1988. His academic publications include 8 books and about 400 peer-reviewed papers in the field of transport processes and thermodynamics. From 1996 to 2001 he was Pro-Rector (Research), Deputy Rector and Pro-Rector (Resources) at Imperial College London.

He is a Fellow of the Royal Academy of Engineering, as well as a Fellow of the Institution of Chemical Engineers, a Fellow of the Institution of Engineering and Technology, and the Institute of Physics. He holds a higher doctorate from Exeter University and honorary degrees from the Universities of Lisbon, Exeter, Loughborough, Southampton and Southampton Solent and is a Fellow of Imperial College London. He was a Council Member of the Engineering and Physical Sciences Research Council of UK until 2011 and has led two reviews of UK science policy in the last four years. He is currently a Visiting Professor at Imperial College London, Instituto Superior Tecnico, Lisbon, and the University of Exeter, Senior Deputy President of the Royal Academy of Engineering from 2011 and its International Secretary; he is the current President of the Institution of Chemical Engineers as well as Chair of the Exeter Science Park Company, a Non-Executive Director of Ilika plc, Chair of the South East England Physics Network, Trustee of the Royal Anniversary Trust, and the Rank Prizes Fund and a member of the Council of Universidade Nova of Lisbon, Portugal . He also serves on a number of International Scientific Committees, has been Chairman of the IUPAC Commission on Thermodynamics and has chaired an expert group for the European Commission on Philanthropic Giving for Research. He holds a number of international awards for his contributions to research in transport processes and was made a Knight Bachelor in the Queen’s Birthday Honours List 2009 for services to Chemical Engineering and Higher Education.