Viscosity and Brownian Motion

Summary

In this combined experiment, the final goal is to determine the size of beads that display Brownian motion in a given solution of glycerol in water. In one part of the experiment, the Brownian motion of the beads is observed with a microscope to determine the diffusion constant, using the ensemble average of the square displacement \( \langle x^2 \rangle \) of the beads. In the other part, the viscosities of several water glycerol mixtures are measured at different temperatures with a capillary viscometer. Both the diffusion constant and the (interpolated) viscosity of the suspension are needed to calculate the bead size.

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1 Introduction

1.1 Viscosity

Thermal conduction, diffusion, and viscosity all belong to the so-called transport properties of a substance. Transport processes take place in gases, liquids, and solids. If, for example, salt is dissolved in a solvent, its distribution becomes uniform by a process we call diffusion. Transport properties, however, are not confined to material flow. Thermal or electric conductivity as well belong to this class of properties. In the case of viscosity, the transported property is the momentum.

The viscosity of a solvent influences the translational and rotational motion of a molecule in solution. Viscosity thus affects for example rates of bimolecular reactions or the sedimentation of macromolecules in an ultracentrifuge. Viscosity is caused by intermolecular interactions between solvent molecules and depends on their size and shape as well as on the temperature.[1]

1.1.1 Definition of viscosity

Consider a liquid that moves in the $x$ direction with laminar flow (Figure 1). Two $xy$ layers separated by a distance $\Delta z$ in the $z$ direction move with velocities that differ by the amount $\frac{\partial v_x}{\partial z} \Delta z$. According to Newton’s law of viscous flow, the frictional force $K_x$ acting between the two layers is proportional to their area $S$ and to the velocity gradient (also called shear rate)

$$K_x = -\eta S \frac{\partial v_x}{\partial z},$$

where $\eta$ is a characteristic property of the substance and is called viscosity coefficient or dynamic tenacity, and sometimes simply "viscosity". Its dimension in the international system of units is kg m$^{-1}$ s$^{-1}$, but it is often given in Centipoise (cp) (1 cp = 10$^{-3}$ kg m$^{-1}$ s$^{-1}$). The measured quantity is often the kinematic viscosity $\nu$, the viscosity coefficient divided by the density of the fluid, i.e. $\nu = \eta/\rho$. A common unit for $\nu$ is Stokes (1 Stokes = 1 cm$^2$ s$^{-1}$).

The frictional force $K_x$ per unit of area is the shear stress

$$\kappa_x = \frac{K_x}{S} = -\eta \frac{\partial v_x}{\partial z},$$
In general, the relationship between shear rate and shear stress has a tensorial form

\[ \kappa = \eta D \]

where \( \tau \) and \( D \) are \( 3 \times 3 \) tensors (matrices), i.e. they are the shear stress tensor and the shear rate tensor, respectively. Shear rate and shear stress are only proportional for **ideal Newtonian fluids**. In this case, plotting \( D \) versus \( \kappa \) gives a straight line with constant slope \( 1/\eta \), i.e. \( \eta \) does not depend on \( \kappa \) (solid line A in Fig. 2). Fluids with low viscosity show approximately this ideal behavior. **Plastic** substances (B) show an analogous linear increase of \( D \) with \( \kappa \), but only once a given threshold \( \kappa_c \) has been reached (yield value). Below \( \kappa_c \) the substance behaves like a solid. **Pseudoplastic** (shear-thinning, C) and **dilatant** (shear-thickening, D) materials, instead, are characterized by non linear behavior, i.e. \( \eta \) depends on the shear rate. Finally, the viscosity can also depend on the time of application of the shear force (thixotrophic or rheopexic).

### 1.1.2 Flow through a tube: Poiseuille’s law

The viscosity measurement of a liquid in a capillary viscometer (Ubbelohde) is based on the description of laminar flow according to Poiseuille’s law. Consider a cylindrical tube of length \( L \) and radius \( R \) (Fig. 3). A laminar flow is characterized by cylindrical layers at radius \( r \) (\( 0 < r < R \)) flowing with velocity parallel to the axis of the cylinder. The velocity increases progressively from the wall of the tube (\( r = R \)), where \( v = 0 \) due to the friction exerted by the wall itself, to a maximum value in the center of the tube (\( r = 0 \)).
The flow can be considered laminar when the so called Reynold number $R^* = \frac{2R\nu}{\eta}$ is smaller than 2000. $R^*$ is a dimensionless number that characterizes the flow behavior. Due to the velocity gradient between neighboring cylindrical layers, the force experienced at a distance $r$ from the center of the tube is

$$K_{\text{inner}} = -\eta \frac{\partial v}{\partial r} |_{r=2\pi L r}$$

where $2\pi L r$ is the surface at the inside of the cylinder. At the outside of the cylinder, at radius $r + \Delta r$

$$K_{\text{outer}} = \eta \frac{\partial v}{\partial r} |_{r+\Delta r} 2\pi L (r + \Delta r) = 2\pi \eta L \left( \frac{\partial v}{\partial r} |_{r} + \Delta r \frac{\partial^2 v}{\partial r^2} |_{r} \right) (r + \Delta r)$$

The forces at the inner and outer surface of the cylinder have opposite signs, because they move in opposite directions relative to their adjacent layers (the liquid is slower outside and faster inside). Under steady flow conditions, the sum of these shear forces must be compensated exactly by the external force driving the motion of the fluid, as for example a pressure gradient between the two ends of the tube:

$$F_{\text{ext}} = (p_1 - p_2) \Delta S = 2\pi r \Delta r (p_1 - p_2)$$

Balancing all forces and neglecting the $\Delta r^2$ terms leads to the differential equation:

$$\frac{\partial v}{\partial r} + \frac{\partial^2 v}{\partial r^2} = \frac{p_1 - p_2}{L \eta}$$

with the general solution (you can check by differentiation):

$$v(r) = \frac{p_1 - p_2}{4\eta L} r^2 + A \ln r + B$$

The constants $A$ and $B$ are determined by the conditions that a) the velocity is zero at the walls of the tube, $v(R) = 0$ and b) the maximum velocity is found at the center of the tube, so $\partial v/\partial r |_{r=0} = 0$. We thus obtain the final result:

$$v(r) = \frac{1}{4\eta L} (p_1 - p_2) (R^2 - r^2)$$

The velocity $v(r)$ has a parabolic profile with maximum at the tube center. By integration over all cylindrical layers it is possible to determine the total volume of liquid $\Delta V$ that flows through the tube per time unit $\Delta t$:

$$\frac{\Delta V}{\Delta t} = \int_0^R v(r) 2\pi r dr = \frac{(p_1 - p_2) R^4 \pi}{8 \eta L}$$

According to Eq. 9, by measuring the time needed by a given volume of liquid to flow through a tube of known radius and length, the viscosity $\eta$ can be determined. Indeed, the capillary viscometer is based on this result, also known as the Hagen-Poiseuille equation. The tube of the capillary viscometer is kept in vertical position, and the driving force moving the liquid through it is the liquid’s own weight. This means that the pressure
gradient at the two ends of the tube is the hydrostatic pressure determined by the height of the liquid column between the top and the bottom meniscus. For a height $h$, the gradient is given by $p_1 - p_2 = \rho gh$, where $g$ is the gravitational acceleration. By applying Eq. 9, the kinematic viscosity becomes

$$\nu = \frac{\eta}{\rho} = \Delta t \left( \frac{\pi R^4}{8 L \Delta V} g h \right) = K_{UV}$$

where all the quantities appearing within brackets are known constants and parameters depending on the geometry of the set-up. They can be reduced to one characteristic constant of the instrument, $K_{UV}$.

#### 1.1.3 Flow resistance of a falling sphere

A sphere of mass $m$, and radius $r_0$ falling in a fluid of density $\rho$, experiences the gravitational force $G$ and a frictional force $K_x$ in the opposite direction. The origin of the frictional force is the velocity gradient between the fluid at rest and the fluid dragged along with the sphere. With equation 1 and $\partial v_x/\partial r \approx v_x/R$, we can estimate

$$K_x \approx -\eta 4 \pi r_0^2 \frac{v}{r_0} = -4 \pi \eta v_x r_0$$

The frictional force is thus proportional to the velocity $v_x$ and the viscosity $\eta$. An exact calculation yields

$$K_x = -6 \pi \eta v_x r_0,$$

known as Stokes law. On the other hand, if $\rho_0 = \frac{m}{4/3 \pi r_0^3}$ is the density of the sphere, the gravitational force is

$$G = \frac{4}{3} \pi R^3 (\rho_0 - \rho),$$

Eventually, both forces are equal and the sphere falls with constant velocity $v_\infty$, which is obtained by equating 12 and 13:

$$v_\infty = \frac{2}{9} \frac{R^2 g}{\eta} (\rho_0 - \rho)$$

Hence, by measuring $v_\infty$, the viscosity $\eta$ can be determined, when all the other parameters are known. Equation 14 is the basis of the falling sphere viscometer.

#### 1.2 Brownian Motion

The English botanist R. Brown discovered in 1827 with the help of a simple microscope, that pollen granules (Clarkia Pulchella) floating in water exhibited a perpetual irregular movement. The particles moved independently of each other in an entirely random manner. Brown was able to show that this movement was not biologically animated when he found that other inorganic materials such as sulfur, volcanic ash, and even fragments of the Sphinx, pulverized (0.8-1.3µm) and dissolved in water also demonstrated the same, now called Brownian motion. Indeed, all microscopic particles in a liquid, such as
molecules or sand / dust particles, are in a constant thermal motion. Nearly eighty years later (1905) A. Einstein devised a theoretical explanation, described by the interaction of a small particle with a heat bath. A full interpretation of Brownian motion was not completed however until J. Perrin (Nobel Prize 1926), who showed that the movement is caused by through collisions between particles and the molecules of the surrounding liquid.\[2\] Perrin’s experiments indicated, in conjunction with Einstein’s theory, a direct proof of the existence of molecules and their random, thermal motion. This motion influences all different kinds of transport and diffusion processes.

In this laboratory experiment, you will observe the Brownian motion of latex beads with the aid of a microscope and a video camera, and collect enough data such that the measurements are statistically relevant. From the data, you will determine the size of the particles from their diffusion coefficients. In the analysis, you will test the equivalence of two statistical metrics, the ensemble average and time average.

In biochemistry simple statements about the form and size of macromolecules can be inferred from the diffusion coefficient. For example, the molecular weight may be estimated of the diffusion coefficient, or if the molecular weight of the particle is already known, it is possible to determine the shape or the hydration volume.

1.2.1 Diffusion

A microscopic theory on the Brownian motion of suspended particles would have to describe the dynamics of the surrounding solvent molecules. It is possible however to construct physically meaningful statements about Brownian motion based on the diffusion properties of the suspended particles. This is possible because the interaction of the microscopic solvent molecules with the suspended particles leads to their macroscopic diffusion.

Consider a liquid in which a quantity of macroscopic particles is suspended. At a time \( t = 0 \), all \( N \) particles are concentrated in one place. Due to diffusion, which decreases the concentration across a gradient, the particles diffuse apart over time. For simplicity, let us consider only the one-dimensional diffusion along the \( x \)-coordinate. The concentration of the particles as a function of time can be described according to Fick’s Law:

\[
\frac{\partial n(x,t)}{\partial t} = D \frac{\partial^2 n(x,t)}{\partial x^2}.
\]

(15)

Here, \( D \) is the diffusion coefficient of the particles, and \( n(x,t) \) is the particle number density (number of particles per unit length) at position \( x \) and time \( t \). Solving this differential equation yields

\[
n(x,t) = \frac{N}{\sqrt{4\pi Dt}} \exp \left[ -\frac{x^2}{4Dt} \right],
\]

(16)

where we have used the following boundary conditions:

\[
n(x = 0, t = 0) \to \infty, \quad n(x \neq 0, t = 0) = 0, \quad n(x \neq 0, t \to \infty) = 0.
\]
1.2.2 Mean square displacement

The mean square displacement $\langle x^2 \rangle(t)$ is the mean of the squares of all the shifts in $x$-direction, which $N$ particles have experienced between time zero and time $t$. When $N$ is sufficiently large, this quantity can be calculated as

$$\langle x^2 \rangle(t) = \frac{\int_{-\infty}^{\infty} x^2 n(x,t)dx}{\int_{-\infty}^{\infty} n(x,t)dx}.$$  \hfill (17)

By replacing $n(x,t)$ with the expression given in equation Eq. 16 and computing the integrals of the Gaussian functions

$$\int_{0}^{\infty} x^2 \exp \left[ -\alpha x^2 \right] dx = \frac{1}{4} \sqrt{\frac{\pi}{\alpha^3}} \quad \text{and} \quad \int_{0}^{\infty} \exp \left[ -\alpha x^2 \right] dx = \frac{1}{2} \sqrt{\frac{\pi}{\alpha}},$$  \hfill (18)

we obtain

$$\langle x^2 \rangle(t) = 2Dt.$$  \hfill (19)

1.2.3 Time and ensemble average

$\langle x^2 \rangle$ is an ensemble average because a family of $N$ particles is averaged at particular time $t$. Since for this experiment it is easier to observe a single particle over a long time period, here we introduce the time average value. Instead of a large number of particles at time $t$, we consider only the position of a single particle at constant time intervals: $\Delta t$, $2\Delta t$, $3\Delta t$, ..., $M\Delta t$. The position of the particle is observed at each time interval to collect a family of coordinates from which a series of displacements as a function of time (discretized as multiples of $\Delta t$) can be computed: $x_1 - x_0$, $x_2 - x_1$, $x_3 - x_2$, ..., $x_M - x_{M-1}$. By taking the square of each displacement and averaging, the mean square displacement at time $\Delta t$ is obtained

$$\overline{x^2}(\Delta t) = \frac{1}{M} \sum_{i=1}^{M} (x_i - x_{i-1})^2.$$  \hfill (20)

This is now a time average, where $M$ is the number of measurements over time intervals $\Delta t$.

Now we make the assumption that time average and ensemble average are equivalent (the validity of this assumption is discussed in many physical chemistry and statistical mechanics textbooks\cite{3}). Based on this assumption and on the fact that the motion of the suspended particles is due to the molecular motion of the surrounding fluid, it is possible to determine the relationship between the displacement of the beads and the diffusion coefficient in the liquid using

$$\overline{x^2}(\Delta t) = 2D\Delta t.$$  \hfill (21)

1.2.4 The Stokes Einstein equation

The link between diffusion, governing the motion of microscopic objects, and viscosity, a macroscopic property of fluids, was made by Albert Einstein in his doctoral thesis\cite{4} (University of Zurich, 1905). Making use of Stokes law (equation 12), he showed that
the diffusion coefficient $D$ of spherical particles of radius $r_0$ in a liquid of viscosity $\eta$ at temperature $T$ is given by

$$D = \frac{k_B T}{6\pi \eta r_0},$$

(22)

where $k_B$ is the Boltzmann constant and $\eta$ is the viscosity of the liquid without the particles.$^{[5,6]}$

## 2 Experiment

You will observe and analyse Brownian motion of latex beads in a water glycerol mixture and determine the diffusion constant $D$. Independently, you will measure the macroscopic viscosity of the water-glycerol solution at different temperatures. If both $D$ and $\eta$ are known, the Stokes-Einstein equation 22 allows you to deduce the bead diameters with a proper error estimate.

### 2.1 Viscosity Measurements

There are six Ubbelohde viscometers in the laboratory, which allow you to measure the kinematic viscosity of Newtonian liquids relative to a calibration substance. They are coupled to two independent thermostats, so you can make several measurements simultaneously, and need clean the apparatus only at the end. Each viscometer has a different characteristic constant $K_{UV}$ and has to be calibrated independently.

#### 2.1.1 Calibration of the viscometer

The characteristic constant of the instrument, $K_{UV}$, can be determined by calibration with a liquid $L_0$ of known viscosity $\eta_{L_0}$. It is important that the chosen calibration
liquid is sufficiently similar to the liquid for which the viscosity has to be measured. The experimental conditions (temperature) need also to be similar. Knowing $\eta_{L_0}$ and the density $\rho_{L_0}$ of $L_0$, the characteristic constant can be calculated using Eq. 10, i.e.

$$K_{UV} = \frac{1}{\Delta t \rho_{L_0}} \eta_{L_0}.$$  \hspace{1cm} (23)

2.1.2 Procedure

- Make yourself familiar with the instrument and the thermostat. Notice that all measurements have to be carried out at a stable temperature, after the whole apparatus has equilibrated.

- Once the target temperature has been set, the thermostat starts cooling or heating the circulating water. When the water has reached the target temperature wait another five minutes for the full thermal equilibration of the experimental set-up.

- Before filling any new solution into the viscometer, remove completely the previous one and clean and dry the capillary. Do not exceed the maximum volume of liquid that can be filled in.

- Prepare a mixture of glycerol in distilled water, identical to the one used in the Brownian motion measurements. Note the exact volumes used. Be sure to mix properly. Fill the (clean and dry!) viscometers and flush the capillaries a couple of times, without taking any data. You can use a water pump to help you to raise the liquid in the capillary tube.

- When the temperature is constant (read and note!), measure the time $\tau$ needed for the fluid to flow in the capillaries.

- To speed up data acquisition, measure simultaneously in the three different viscometers.

- Record data at at least four different temperatures $T$, within the temperature range of the Brownian motion experiment (between 20° and 30° C).

- Clean the viscometers and repeat the measurement with a substance of known viscosity for calibration (at a single temperature for which its viscosity is known).

2.2 Brownian Motion

2.2.1 Sample preparation

- You will be provided with a stock suspension of latex beads of unknown diameter in a water. Make a water/glycerol mixture (at a ratio specified by the assistant) both with and without beads. Shake the suspension vigorously and remove 250 $\mu l$ of solution and dilute with 1 ml of the bead-free mixture.
• To make a measurement cell, fill one or two drops of the prepared suspension into the small sink of the special microscopy slides and seal the cell with a thin glass cover slide. Use vacuum grease for sealing if necessary and remove solution in excess.

• Inspect the slide under the microscope and view the Brownian motion of the beads. The concentration of beads should be high enough for a single movie to render the trajectories of at least five beads at the same time. On the other hand, a too high concentration makes it difficult to track single beads. If the concentration of the beads is too low or too high, make a new solution and cell.

Conceptual questions:

• Is it possible to discern Brownian motion from other types of motion, and what is the probable cause of these motions?

• Why is there a heat filter glass between the slide and the light source?

2.2.2 Calibration

The software we will use for data analysis requires inclusion of a few frames showing the snapshot of the calibration marks.

• Focus on the calibration marks such that you can see 2 large hashes and the small hashes in between. The distance between the large hashes is 100 \( \mu m \), the distance between medium hashes is 50 \( \mu m \) and 10 \( \mu m \) between the small ticks.

• In the capture options section of the capture tab, take a snapshot (camera icon) of the calibration lines.

2.2.3 Data Acquisition

• Open the video capture application Power Director. Switch to the capture tab and record several movies of Brownian motion. The clips will then become available under the editing tab.

• When recording the movies, be very careful not to bump into or jostle the microscope. Jumps caused by this motion differ from Brownian motion and can be identified by the concerted movement of all of the beads in the same manner. This type of motion should be edited out.

• Be sure that your measuring chamber is sealed well. Any leakage will be seen as directed motion of the beads. Even if it looks good at first sight, you should check it by playing the first recorded movie fast forward. In case of a bad seal, prepare the measuring chamber again.

• Do not forget to measure and note down the temperature of the sample under the microscope. Use the same thermometer as in the viscosity measurements.
2.2.4 Producing videos

Under the edit tab, you can edit together clips of Brownian motion into movies.

- Movies should be about 60–90 seconds long in total. It is not necessary to go for much longer times.
- The filmed motion should be one continuous video, with no concerted, non-Brownian motion, such as that caused by a bump to the microscope. If there is such motion in a recorded clip, edit it such that only motion before or after is used in this particular movie, but do not combine two non-continuous clips together.
- Include a 1–2 second clip of the calibration marks at the beginning or the end of the movie for data analysis later.

Under the production tab, produce the movie. Use the default settings to make an AVI movie. Repeat these steps to make at least 3 movies that are suitable for analysis and allow you to track at least 15 different beads. Record additional movies if necessary.

3 Data analysis

3.1 Movie analysis

- Import the movies into the tracker analysis software. This can be done on a computer in the lab, or on your own computer. You can find a free download at http://physlets.org/tracker/.
- Calibrate the distances using the first frames of the movie according to the instructions provided by the application. There are extensive help files and descriptions included with the program.
- Track the location of a particle undergoing Brownian motion for as long as possible in the movie. Repeat for as many other visible particles as possible within the movie, and for the other movies.
- Export the time and location data to a spreadsheet or analysis program for further analysis. Do not make the time steps too small (0.2-0.4 seconds).

3.2 Trajectory analysis

- Import the time and position \((x, y)\) of your recorded trajectories into a spreadsheet program (or other suitable program). Include in your report an illustrative plot of the \(y\) versus \(x\) position from at least one of these trajectories.
- Calculate the mean square displacements \(\Delta x^2\) and \(\Delta y^2\) in the \(x\)- and the \(y\)-directions as a function of time interval \(k \Delta t\), \((k=1, 2, 3\ldots)\). One way to do this is shown in the table below, where the displacements have been measured along the \(x\) direction. Do not chose \(\Delta t\) too small or too large! If the table is properly constructed, it should be possible to analyze all of the trajectories in both \(x\) and \(y\) directions using the same table of formulas.
• In the same table, determine the standard errors \( \sigma_{\Delta x^2} \), given by the standard deviations of the data corrected by the number of rows \( n \) used for averaging (divide by \( \sqrt{n-1} \)). There should be at least 10 points per data set (time interval).

• Make a new table (in Origin), where you collect \( \Delta x^2 \) and \( \Delta y^2 \) and their uncertainties for all trajectories. The first column should contain the different time intervals \( k \Delta t \). Plot the mean square displacements as a function of time interval including the standard errors as error bars. Show linear fits of the data.

• Produce a plot illustrating the combined result from all data points from your best data sets (please explain your criterion in selecting or omitting data) with error bars. Perform a linear fit of the data, using the error bars. From the slope, determine the diffusion coefficient \( D \) and its error.

### 3.3 Viscosity

In order to determine the viscosity \( \eta(T_{Br}) \) at the temperature \( T_{Br} \) of the Brownian motion experiments, we interpolate the data recorded with the flow viscometers. For most fluids the viscosity decreases exponentially with increasing temperature:

\[
\eta = A e^{E/k_B T} \tag{24}
\]

which can be rewritten in logarithmic form as

\[
\ln \eta = \ln A + \frac{E}{k_B} \frac{1}{T}. \tag{25}
\]

By plotting \( \ln \eta \) as a function of the inverse of the temperature, regression analysis can be applied to determine the two parameters \( \ln A \) and \( E/k_B \), which are the intercept and the slope, respectively. The viscosity at any intermediate temperature can then be deduced with an appropriate error. Proceed as follows:

• Compute all viscosities according to

\[
\eta(T, i) = \rho K_{i}^{UV} \tau(T, i) \tag{26}
\]

where different \( i \) stand for different viscometers. For the calibration constants \( K_{i}^{UV} \) use equation 23 with the standard deviation of your calibration times.
• For the density $\rho$ of the water glycerol mixture assume that it only depends on the volume fraction:

$$\rho = \rho_W \frac{V_W}{V_{tot}} + \rho_G \frac{V_G}{V_{tot}}$$

(27)

. However, you may also determine the density experimentally.

• Calculate the averages $\bar{\eta}(T) = \sum_i^N \eta(T, i)/N$ at each temperature and plot $\ln(\bar{\eta})$ against $1/T$ (in Kelvin!). Use appropriate error bars.

• Perform a linear fit and hence determine $\eta(T_{br})$ with an error bar that takes into account the potential difference between the temperature measurements in the two experiments (microscope and viscometer).

3.4 Final steps

From the diffusion constant $D$ and the interpolated viscosity $\eta(T_{br})$ determine the bead diameter $r_0$ and its experimental uncertainty for both of the latex beads used.

In your report you may want to comment on the following points:

• What is the expected value of the average displacement of a particle over many measurements? Did your data show this?

• What is the relationship between the mean displacements in $x$ and $y$ direction? What about the average square displacements?

• The suspended particles are in a continuous fluctuating motion indefinitely. Consequently, are these particles in a kind of perpetual motion? Could Brownian motion be used for a perpetual motion machine?

• Why can we use the assumption that the time average and ensemble average values are equivalent?

• Discuss the significance of Brownian motion to chemical reactions, biological or physiological systems.

Constants:

$k_B = 1.3806 \times 10^{-23}$ J K$^{-1}$

$R = 8.3145$ J K$^{-1}$ mol$^{-1}$

$N_A = 6.0221 \times 10^{23}$ mol$^{-1}$.

For the viscosity of water/glycerol mixtures see also.$[^7]$

References


