Measuring Velocity Distributions of Viscous Fluids Using Positron Emission Particle Tracking (PEPT)

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Positron emission particle tracking (PEPT) can be used to trace the path of a radioactive particle within opaque fluids in pilot-scale equipment; the method can track particles through several centimeters of metal. PEPT has been successfully used to follow isokinetic tracers in viscous fluids and thus to measure velocity distributions under both isothermal and nonisothermal conditions in pipe flow. The accuracy of the method decreased as the measured velocities increased; the faster the particle traveled, the less accurate its detection. For velocities of up to 0.5 m/s the accuracy of the method was acceptable. Agreement between experimentally measured and theoretical velocity distributions was very good, for a range of fluids and process conditions. As tracer particles are used, there were problems ensuring that all parts of the measurement volume were sampled. This is possible to overcome to an extent by adjusting particle size; 600-μm tracers did not pass within 1 mm from the tube wall, whereas 240-μm particles passed much closer to the boundaries of the flow.
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Introduction

Many industrial processes, particularly in the food and fine chemical industries, involve heat and momentum transfer in opaque fluids with complex rheological properties that are often time dependant. Measuring flow and heat transfer is important because it is often the microstructure, generated as a result of processing, that gives the required properties to the product. Measurement of the local velocity and temperature fields is challenging, however, because of the opaque nature of the material.

A number of methods have been used to measure velocity distributions. Some methods are suitable only for transparent fluids. For example, Bakalis and Karwe (1998) and Chandrasekaran and Karwe (1997) measured velocity distributions in a twin-screw extruder using laser Doppler anemometry (LDA). Because LDA requires optical access to the flow, a Plexiglas window was fitted to the vent port of the extruder. Particle imaging velocimetry (PIV) has also been used [for example, by Pakdel and McKinley (1997)] to measure velocity distributions within viscoelastic fluids. However, because both LDA and PIV are methods that require optical access to the flow, not only the materials used but also the equipment has to be transparent to allow flow materials to be quantified.

Magnetic resonance imaging (MRI) has also been used to measure velocity distributions in opaque systems and in complex fluids, such as in suspensions (Li et al., 1994), in packed beds (Johns et al., 2000), particle–liquid mixtures (McCarthy et

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al., 1997), Couette flows (Hanlon et al., 1998), and in scraped-surface heat exchangers (Wang et al., 1999, 2000). The method is able to make rapid and detailed measurements of flows. Although velocity measurements can be performed in opaque materials, using MRI, the experimental equipment cannot contain metal because of the nature of the measurement technique. This limits the method, thus making it difficult to measure velocity distributions in realistic equipment, such as under heat-transfer conditions.

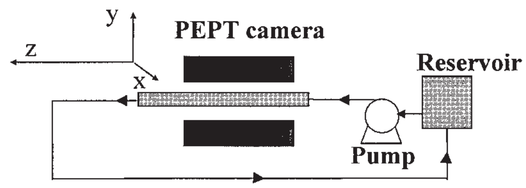
There is a need to develop a method by which velocity distributions can be quantified under realistic conditions. The School of Physics at Birmingham has developed a unique way of following particles and flows in opaque fluids, using positron emission particle tracking (PEPT) (Parker et al., 1993). In this technique a tracer particle ($\leq$600 μm in diameter) is passed through a flow or mixer system. The tracer has been treated so that it is slightly radioactive, emitting positrons that then generate a pair of back-to-back gamma rays on collision with an electron. The two detectors are placed on either side of the system of study and operate in coincidence mode, an "event" being recorded only if the γ-rays are detected by both detectors with a resolving time of 12 ns.

The camera data rate can operate up to 100,000 coincidence events/s, so that acquiring sufficient data to enable tomographic reconstruction of an extended tracer distribution may take many minutes. Use of the camera in this way has therefore concentrated on a range of steady-state situations. Applications have ranged from imaging lubricant distribution within an operating jet engine (Stewart et al., 1987), to mapping the concentration of sand grains in a stirred slurry (McKee et al., 1995).

For PEPT, a single tracer is used whose position and velocity can be followed by detecting the γ-rays and triangulating their position. The radioisotope used (^{19}F) has a half-life of 2 h, so that residual radioactivity is negligible after a few hours. The technique can be used in pilot-scale equipment as the γ-rays can penetrate reasonable thicknesses of metal (up to 10 cm). Many of the detected events are corrupt, for example, because one of the γ-rays has been scattered: location requires acquisition of sufficient events so that the valid ones, whose reconstructed lines essentially meet at one point, can be distinguished. The number of events used depends on various factors, including the tracer speed.

In general a slow moving tracer is located to within $\pm$1 mm, typically 100 times/s. Tracers are made by soaking ion-exchange resin beads in radioactive water; they are then coated in paint to retain the radioactive fluid. PEPT has been widely used to study mixing patterns in powders and soft solids; here a tracer is placed in a mixer and circulated for some time to build up an image of the flow field. The method has been used in a number of processes, such as the flow of powders in rotating drums (Parker et al., 1997) and fluids in stirred tanks (Fangery et al., 2000). Recent work has developed the technique to follow trajectories in flowing systems, such as solid–liquid flows in pipelines (Fairhurst et al., 2001). In this case the tracer was inserted into a large (nonisokinetic) particle and circulated through the process, allowing a set of trajectories to be mapped.

PEPT has not previously been used to study the flow behavior of liquids. Recent development in particle handling, however, has resulted in the production of tracers that are sufficiently small to be essentially isokinetic with the fluid. The objective of this study was to develop and validate a method for velocity measurements in viscous opaque fluids using PEPT. To demonstrate the technique, velocity distributions were measured in horizontal pipes for fluids of known rheological properties, and experimentally measured and theoretically predicted velocity distributions compared. This work is a necessary prerequisite for the study of flows in more complex geometries where such validation would not be possible.



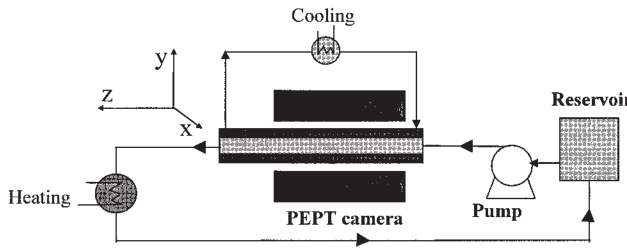


Figure 1. Experimental setup for isothermal (a) and nonisothermal (b) experiments.

Materials and Methods

Experimental equipment

To develop the method a relatively simple apparatus was used. Because the radioactive particle has to remain in the system to measure the velocity distributions, a simple closed-loop system had to be used. Velocity distributions were measured in an aluminum pipe (diameter 19 mm). The apparatus is shown in Figure 1a. To measure velocity distributions under nonisothermal conditions, a tubular heat exchanger (inner tube diameter 37 mm) was used, shown in Figure 1b. In this case, electric heaters were used to bring the temperature of the circulating water to 70°C, and as cooling medium, water cooled in a bath to 5°C was passed through the shell side of the exchanger. In all cases, fluid Reynolds numbers were lower than 10.

Fluid was circulated using a gear pump to minimize the stress applied to the tracer particle: when a centrifugal pump was used the tracer often broke into a number of smaller radioactive particles. Because the volume of the fluid in the system was relatively high compared to the flow rate, the time needed for a particle to recirculate was approximately 2 min. To decrease this time, up to seven particles were introduced to the system so that one was always in the field of the camera. Preliminary experiments used about 70 particle passes. Although the measured velocities agreed well with the theoretical, velocity values were obtained in only a small part of the flow domain (that is, over a narrow band of radii). Later experiments were conducted in which nearly 200 passes were recorded.

Materials

Materials with well-defined rheological properties were used. Two fluids with non-Newtonian rheology were used together with one Newtonian fluid of simple flow behavior:

(1) *Newtonian fluid*: a 37% sucrose solution with viscosity of 0.5 Pa·s.

(2) *Shear thinning fluid:* a 1% aqueous CMC solution, for which $k = 8.5$ $Nm^{-2} \cdot s^{0.55}$ ($n = 0.55$).

(3) *Herschel–Bulkley fluid:* a mixture of 35% sucrose and 1% CMC solution, $k = 9.99$ $Nm^{-2} \cdot s^{0.44}$, $n = 0.44$, $\tau_y = 12$ Nm^{-2}.

In the latter case the constitutive equation is

$$\tau = \tau_y + k\dot{\gamma}^n \tag{1}$$

The rheological properties of the solutions were measured using a cone-and-plate rheometer at shear rates up to 60 s^{-1}.

Estimation of velocity distributions from particle paths

PEPT produces a set of particle trajectories showing how the tracer moved through the equipment. The velocity component along the axis of the tube (V_z) is related to the particle path with the following equation

$$V_z = \frac{dz}{dt} \tag{2}$$

where z is the axial spatial coordinate and t is time.

The PEPT data consisted of locations of the particle (x, y, z coordinates) at various times. In the geometry considered only the axial velocity component (here termed z velocity component) was of importance. This was determined as a function of the radial position in the channel. Using regression analysis a line was fitted to a number of the z locations of the particle vs. time. The slope of the line was the axial velocity component of the particle.

Various methods were used to estimate velocities. All depend on the accuracy of the tracer location. Consecutive positions of the tracer were typically obtained within fractions of a second and differed in position by less than 0.5 mm. Given that the point where an emitted positron meets an electron is not necessarily at the same point as the tracer, there is local scatter in the measured position, leading to significant error in the velocity measured. These points could be identified as rapid "jumps" in the track of the particle with time.

After removing these points a moving average was applied to smooth the particle path. Velocity values were estimated by fitting a line to a number of particle locations. Typically about 15 consecutive particle positions were used. The axial distance traveled was less than 1 mm. In this distance it was assumed that there was no significant change in the radial location of the tracer: if the standard deviation in the radial location was >0.5 mm, velocity values were discarded.

Results and Discussion

Sedimentation velocity of tracers

To measure the velocity field of a liquid using the PEPT method it must be confirmed that the particle is isokinetic with the fluid (that is, the particle follows the streamlines of the fluid), without having an effect on them. The relatively high viscosity and density of the fluids, together with the small diameter of the tracers, suggested that the particle would be isokinetic. The sedimentation velocity of a typical tracer particle was measured in the sugar solution and a 1% CMC solution. The sedimentation velocity of a typical particle in a 37% sugar solution was found to be about 0.001 m/s, that is, 2 orders of magnitude smaller than the axial velocities of interest (which are on the order of 0.1 m/s) and therefore can be neglected. In the case of 1% CMC solution the particle did not sediment at a measurable rate.

PEPT measurements and velocity determination

In Figure 2a a front view of typical PEPT data is shown (that is, the set of x– y coordinates recorded). The particle is clearly confined within a circular geometry. Some data points appear to be outside the pipe, which is not physically possible. This may be attributed to one of two reasons:

(1) The algorithm currently used can track only one particle at a time and sometimes more than one particle was between the cameras. In this case it is possible to obtain positions of the particles outside the pipe. These cases are obvious and are removed during data analysis.

(2) The distance traveled by the positron before destruction is not constant. It is possible that some will travel outside of the pipe to give spurious positions.

One additional problem is that the pipe will never be perfectly aligned to the camera. In Figure 2b the top view (x–z) of the same data set is shown. The pipe was not perfectly aligned with the axis of the cameras, so that particles appear to move at an angle to the axis of the camera. Before final analysis was carried out, the data sets were transformed so that the pipe axes were used, as in Figure 2c, where the squares represent the same data rotated to be aligned with the axis of the pipe.

This was done as follows: a radioactive tracer was placed at 10 to 12 positions on the outer surface of the pipe at two axial positions. The center of the pipe was obtained by fitting the equation of a circle to these points. If the axis of the pipe were perfectly aligned with the axis of the camera there would be no difference in the coordinates of the center at different axial positions. For a typical experiment, the x coordinate of the pipe center changed by approximately 3 mm within the measurement volume, whereas the y coordinate changed about 0.2 mm. The data need to be rotated only along the x–z plane. The equation used was

$$\begin{pmatrix} x' \\ y' \\ z' \end{pmatrix} = \begin{pmatrix} \cos\varphi & 0 & \sin\varphi \\ 0 & 1 & 0 \\ -\sin\varphi & 0 & \cos\varphi \end{pmatrix} \begin{pmatrix} x \\ y \\ z \end{pmatrix} \tag{3}$$

where x, y, and z are the locations recorded from the camera; x', y', and z' are the rotated coordinates along the axis of the pipe; and φ is the angle between the axis of the pipe and the camera, which is calculated from the following equation

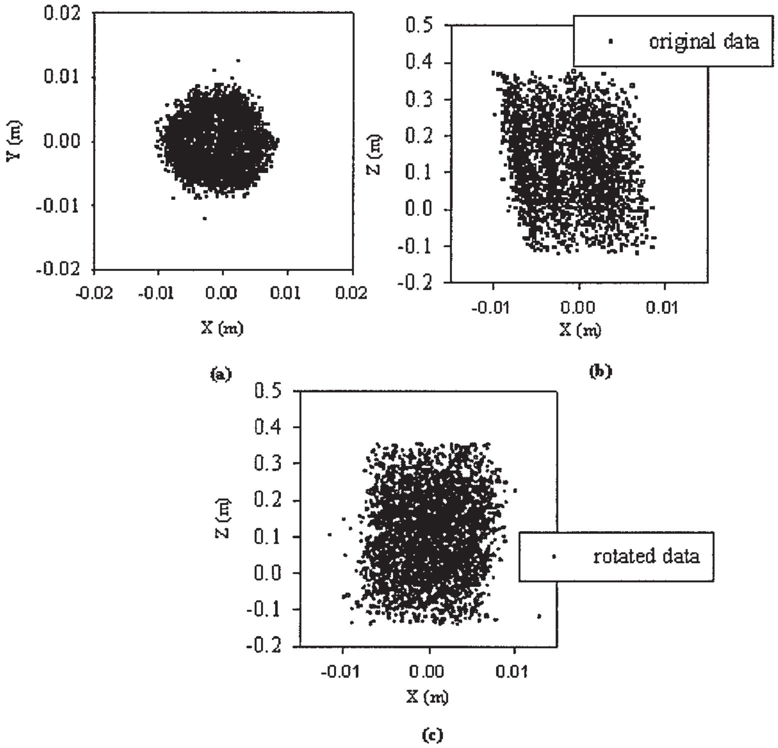


Figure 2. Side (a) and top (b) and (c) views of typical PEPT data in a straight pipe under isothermal conditions.

$$\tan \varphi = \frac{x_{c1} - x_{c2}}{z_{c1} - z_{c2}} \tag{4}$$

where x_{c1} and z_{c1} and x_{c2} and z_{c2} are the x and z coordinates of the center at the two axial positions. The rotated data were then used to estimate the velocity distributions.

One of the differences between the information gathered using PEPT and that using fluid tracers (that is, the conventional dye test used to determine a residence time distribution) is that the circulated tracers may not provide information on the total flow behavior of the fluid. This will occur because some fluid paths are so rare that they will not be followed by the finite number of tracer paths through the equipment. In addition, there will be a limit on the distance that a tracer of finite size can approach the boundaries of the flow. For this reason, only distributions of passage time rather than true residence time distribution (RTD) can be measured using the tracer data directly (for more detail, see Lareo et al., 1997).

For measurement of a velocity profile, however, this limitation is not important, providing that a full range of radii is mapped out by the tracers. It is thus necessary to compare the theoretical and actual number of tracers at different radii. If infinitely small particles are randomly distributed across the cross section of the tube, the probability density function $f(r)$ of a particle being at a radius r is given by

$$f(r) = \frac{2r}{R^2}\, dr \tag{5}$$

In Figure 3 the probability density function of a particle passing at a radius r predicted from Eq. 5 is compared with experimentally observed particle radii. The agreement between the experimentally measured and theoretically predicted probabilities is quite close up to about 8 mm. The largest radius the tracer can physically reach is the maximum radius minus half the tracer diameter. This represents a limitation of the method: experimentally, the number of tracers identified at high radius is small, as shown in Figure 3.

The effect of the number of points used in the regression analysis on the calculated velocity values was examined (Bakalis et al., 2003). If there was no experimental error the velocity distributions would not depend on the number of points used in the regression. Because there is experimental error, sufficient points must be taken to ensure that the velocity distributions are independent of the number of points used in the regression analysis. Statistical analysis (Bakalis et al., 2003) found that if more than 15 points are used in the regression analysis no differences occur in the statistics of the velocity distributions, and thus 15 points were used throughout.

Results for velocity profiles

A series of experiments were carried out to confirm the accuracy of the technique.

Newtonian Fluid. In Figure 4a velocity distributions for a 37% sucrose solution in a straight pipe for two flow rates are shown. At a pump speed of 700 rpm the measured volumetric flow rate was 6.65×10^{-5} m^3/s, whereas at 1000 rpm it was 7.55×10^{-5} m^3/s. For both flow rates the experimental and theoretical velocity distributions agreed very well (within 10%). At higher flow rates there is more scatter in the velocity distributions, which would be expected, given that tracers are located more accurately when they move at a lower speed.

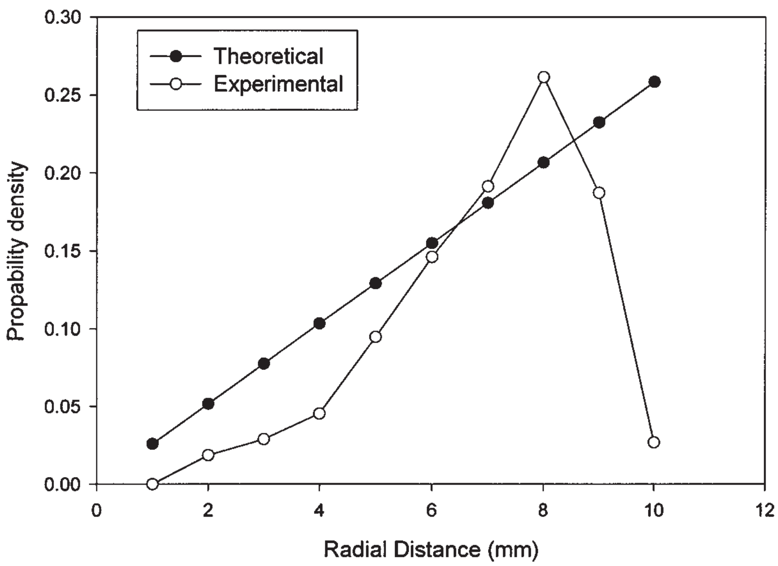


Figure 3. Experimental and theoretical probability of a particle passing at a specific radial distance in a shear thinning solution.

Shear Thinning Fluid. In Figure 4b velocity profiles measured for the same two flow rates for a 1% CMC solution, a shear thinning fluid, are shown. As with the Newtonian case, velocity distributions agreed well with the theoretical predictions; agreement is especially good closer to the center of the pipe. It is important to note that in both cases no particles passed within 1 mm from the pipe wall so that it is not possible to confirm the velocity in the near-wall region. This represents another limitation of the method: particles pass through the fluid at randomly determined radii. Few data points are found at the center, where the flow area is low, and at the outside region, where the velocity is low. Particle migration in shear flows has been determined in some cases (Lareo et al., 1997), although this was not detected within the camera.

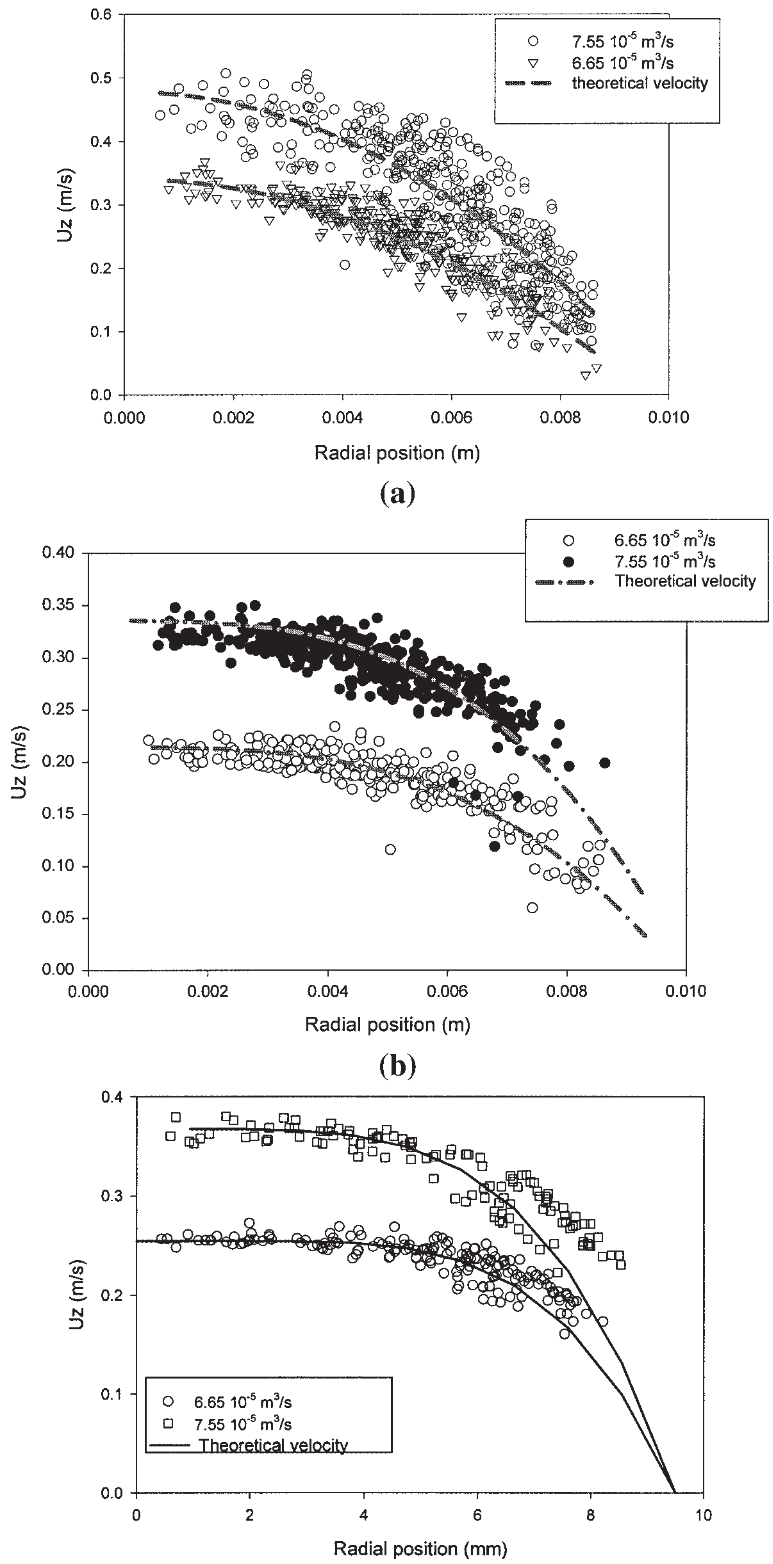
Herschel–Bulkley Fluid. In Figure 4c velocity distributions measured in a sucrose–CMC solution are shown for the same two flow rates. The solution used is a Herschel–Bulkley solution and exhibits a yield stress. Experimentally measured velocity distributions agreed very well with the theoretically predicted values. The limitation of the method observed when using CMC was also seen here; no particles passed within 1.5 mm of the outer radius of the pipe. As previously mentioned, depletion of the tracers from the boundaries of the flow might be the result of particle migration. It has been shown (Fryer et al., 1999) that particle migration depends on the particle size: the larger the particle, the faster the migration.

To investigate the effect of the tracer size on the radii across which velocity distributions were measured, a smaller tracer was used. Figure 5 shows velocity distributions measured for two particle sizes for a sugar–CMC solution. The nominal diameter of the smaller particles was 240 microns, although after coating the particle diameter increased to about 280 μm. The smaller particle was able to give velocity measurements closer to the pipe wall, whereas the agreement with theoretical velocity profiles was better closer to the boundaries. These results indicate that the particles used should be as small as possible; however, there are practical limitations that prohibit the use of particles smaller than 240 μm. To keep the particles active for 3 h (the time scale of an experimental run) they needed to be coated; with particles smaller than 280 microns this is very difficult. Work is under way to develop better and smaller tracers.

The above results indicated that PEPT can be used to measure velocity distributions in viscous fluids. The limitations and the accuracy of the method have been explored using isothermal systems in which the velocity distribution can be obtained analytically. The particular advantage of PEPT is that it can detect tracers through metal, under process conditions. To explore the capabilities of PEPT further, therefore, velocity distributions were measured within a stainless steel concentric tubular heat exchanger with an internal diameter of 37 mm. A mixture of 250- and 600-μm tracers was used.

Theoretical velocity distributions in the tube for steady flow were estimated using a commercial finite-element program (FIDAP), assuming a constant wall temperature. The program solved the continuity, motion, and energy equations (Bird et al., 1964, 318–319). The equations are coupled because viscosity depends on temperature, which changes both axially and radially along the heat exchanger. The rest of the physical properties were considered independent of temperature. The dependency of viscosity on temperature was obtained from Pereira et al. (1993). Inlet and boundary conditions were: (1) parabolic velocity profile in the entrance region and a uniform temperature of 70°C, (2) zero velocity and 5°C temperature at the wall. The fluid entered the heat exchanger through sufficient length of straight pipe, with diameter equal to that of the tubular heat exchanger, to ensure the parabolic boundary condition used in the simulation was valid.

In Figure 6 the velocity field when using 1% CMC solution is shown. Velocity distributions were averaged over an axial distance of 5 mm, in the middle of the tubular heat exchanger (that is, at an axial distance of 30 cm). To obtain adequate data experiments were repeated three times. Experimentally measured velocity distribution agreed very well with the theoreti-



(c)

Figure 4. (a) Velocity distributions in a pipe for different flow rates for a sugar solution (Newtonian); (b) velocity distributions in a pipe for different flow rates for a 1% CMC solution (shear thinning); (c) velocity distributions in a pipe for different flow rates for a CMC–sucrose solution (Herschel–Bulkley).

cally predicted from FIDAP. Both the isothermal and heat-transfer simulations are shown in the figure, and it can be seen that the experimental data are closely predicted by the nonisothermal simulation. Using a mixture of particles, it was possible to measure velocity distributions, even close to the boundaries of the flow. Better agreement might be achieved with a

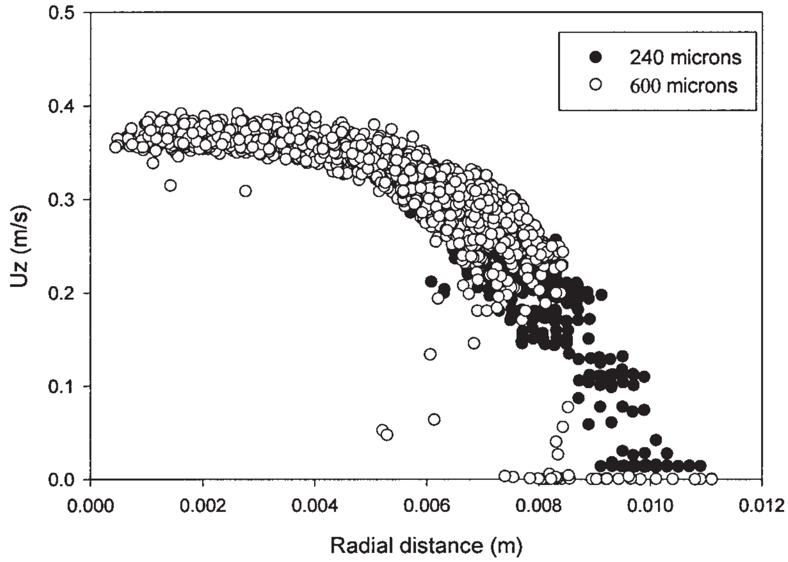


Figure 5. Velocity distributions in a pipe for a 1% CMC 35% sugar solution (Herschel–Bulkley fluid), under isothermal conditions using two different tracers with respect to size.

more sophisticated model, but the approximations made in the simulation make it simpler to run.

Applicability of the method

The PEPT technique clearly allows velocity profiles to be measured in the cases shown above; in all cases there are some outliers, but they do not affect the method. The method is not ideal; alternatives such as MRI or ultrasonics can measure whole velocity profiles, and do not require the use of tracers. However, the technique does not require physical contact with the equipment, and can visualize flows through metal. The method is applicable here because of (1) the high viscosity of the fluid that makes the tracers isokinetic to the fluid; and 2) the ratio of the tracer diameter to that of the tube, which minimizes the impact of the tracer on the velocity profile. Design of tracers is obviously critical; the method requires that sufficient events are detected by the camera, which limits the tracer size and the velocity that can be measured accurately, and that the tracer is isokinetic and does not disturb the flow field. More work needs to be done to determine the upper limit of velocity and the degree to which the tracers need to be isokinetic. It seems likely that if the sedimentation velocity of the tracer is less than 1% of the flow velocity, and the particle size is less than 5% of the tube diameter, then results will be sufficiently accurate.

Conclusions

PEPT allows the measurement of tracer position and velocity through several centimeters of metal. The PEPT technique has

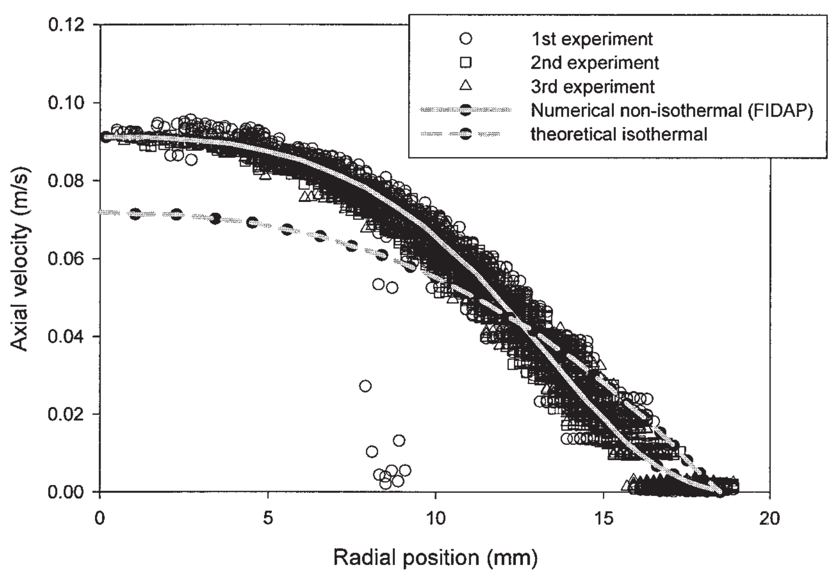


Figure 6. Velocity distributions in a tubular heat exchanger for a 1% CMC solution (shear thinning fluid), under nonisothermal conditions for three sets of experiments, compared with FIDAP simulation.

been successfully used to measure velocity distributions of fluids with various rheological properties, using isokinetic tracers. Velocity profiles were identified by taking an average of 15 readings of position.

Velocity distributions were measured in an aluminum pipe for model transparent and nontransparent fluids. The agreement of the experimentally measured and theoretically predicted velocity distributions was good. Velocity profiles were also measured within a simple concentric-tube heat exchanger; the measured velocity profile is very close to that predicted from computational simulation. A range of tracer sizes were used: whereas 600-μm tracers did not pass within 1 mm from the tube wall, 240-μm particles passed much closer to the boundaries of the flow. The experiments have shown the feasibility of using flow-following tracers to track flows using PEPT; further work is under way to study geometries of greater complexity and industrial significance.

Acknowledgments

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Notation

k = consistency index of shear thinning fluid, $Nm^{-2} \cdot s^n$
L = length of the pipe, m
n = flow index of shear thinning fluid
r = radial distance, m
R = radius of the pipe, m
t = time, s
V = velocity, ms^{-1}
x = x coordinate, m
y = y coordinate, m
z = z coordinate, m
ΔP = pressure drop, Nm^{-2}
τ_y = yield stress, Nm^{-2}
μ = viscosity of a fluid, $Nm^{-2} \cdot s$

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