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Velocity measurements in complex flows of non-Newtonian fluids

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Abstract

Experimental methods for making quantitative measurements of velocity fields in non-Newtonian fluids are reviewed. Techniques based on light scattering spectroscopy - laser Doppler velocimetry and homodyne light scattering spectroscopy, techniques based on imaging the displacement of markers - including particle image velocimetry and molecular tagging velocimetry, and techniques based on nuclear magnetic resonance imaging are discussed. The special advantages and disadvantages of each method are summarized, and their applications to non-Newtonian flows are briefly reviewed. Example data from each technique are also included.

Keywords : laser doppler velocimetry, particle image velocimetry, NMR imaging

1. Introduction

Velocity measurements in complex flows of non-Newtonian fluids are important for a number of reasons. In processing flows, these measurements may provide the most direct method of determining conditions under which the flow becomes unstable, how efficiently materials are being mixed, where "dead zones" are occurring that may lead to heat or mass transfer problems, and where high shear rate regions exist that may lead to shear degradation. In terms of progress in predicting and understanding non-Newtonian flows, quantitative flow measurements on benchmark problems are vital. Any improved constitutive model for viscoelastic fluids must ultimately be judged by its ability to predict a measured complex flow field. Similarly, advancements in numerical simulation techniques must be tested against measurements of velocity fields in nontrivial flows.

Non-Newtonian fluids typically present their own set of unique challenges to the experimental fluid dynamicist. They are often extremely viscous, elastic, and opaque. The high viscosity can lead to significant viscous heating, and can result in temperature and refractive index gradients that will interfere with some optical methods. The high elasticity can lead to steep stress and velocity boundary layers and to long, thin wakes in flows past obstacles. This makes spatial resolution of the measurement technique important. It also prevents the use of any invasive technique for flow measurement, such as hot wire anemometry, since the probe itself will result in a non-trivial perturbation to the flow. Hot wire probes are also adversely affected by viscous dissipation around the probe, which can affect the heat transfer characteristics and interpretation of the results. Finite aspect ratio or "end" effects can propagate significantly further into viscoelastic flows than Newtonian flows, and entry lengths are also frequently much longer. Thus in using any technique that measures only two-dimensional flows or that averages over a "neutral" dimension, special care must be taken to insure that the aspect ratio of the flow is suitably high. Further contraints apply to non-Newtonian fluids that are opaque or contain large numbers of particles; for these fluids, optical techniques fail and one must use nuclear magnetic resonance imaging techniques.

Fortunately, the number of tools for velocity measurement have expanded considerably in recent years. Here, we present a brief review of the main techniques available for measuring velocities in non-Newtonian fluids. Techniques based on light scattering spectroscopy, on direct imaging of tracer displacements, and on nuclear magnetic resonance imaging are briefly described. Special advantages and considerations – whether the technique allows for global or pointwise velocity measurements, whether special seed particles or contrast agents must be added, whether the fluid must be optically transparent – for using each technique are discussed. Finally, applications of each technique to non-Newtonian fluids are briefly summarized.

2. Techniques based on light scattering spectroscopy

A small particle in a flowing liquid will scatter light from an incident laser beam. The laser beam and the optics for

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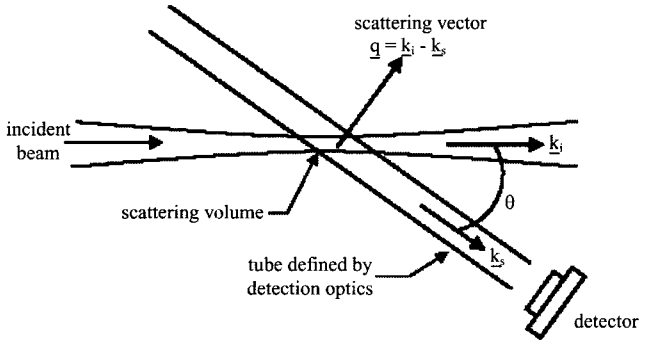


Fig. 1. Geometry for light scattering spectroscopy. The incident light wave vector is $\underline{k}_i$, the scattered light wave vector is $\underline{k}_s$, and the scattering vector q is defined as $\underline{k}_i - \underline{k}_s$. The scattering volume is defined by the incident light beam and the detection optics.

collecting the scattered light define a measuring volume (see Fig. 1). The time dependent light scattering involves time scales associated with the average velocity as the particle passes through this volume, the diffusion of the particle, and a time scale associated with the velocity gradient. (For anisotropic particles and unsteady flows, additional time scales related to particle rotation and the variation of the flow may also be important.) There are two types of light scattering used in flow measurements: homodyne and heterodyne. The spectrum of the scattered light mixed with the incident light, the heterodyne correlation function, is typically dominated by the time scale associated with the mean velocity of the scatterers in the measuring volume. This heterodyne light scattering is used in laser Doppler velocimetry to make pointwise measurements of the velocity field. In the homodyne mode, the scattered light is mixed with itself at the photodetector, and the effect of the mean velocity on the correlation function is negligible provided the other time scales are sufficiently short.

When the time scale associated with the velocity gradient is much smaller than that for diffusion, homodyne light scattering (or photon correlation spectroscopy) can be used to directly measure the velocity gradient at a point in the flow. These two techniques are discussed below. If the fluid is quiescent, information about the diffusion of particles or molecules is obtained from the time dependence of the scattered light; this is the most frequent use of dynamic light scattering or photon correlation spectroscopy. This conventional use of dynamic light scattering to obtain diffusion coefficients (which may be related to hydrodynamic radii through the Stokes-Einstein equation), rather than information about flow, is not discussed in this review.

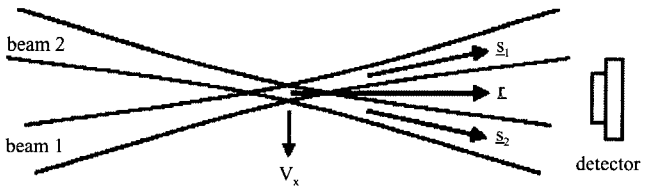
2.1. Laser doppler velocimetry

Laser Doppler Velocimetry (LDV) is a non-invasive, pointwise technique for measuring the local velocity in a flowing fluid. The high spatial and temporal resolution allow its use in a broad range of situations, from capillary blood flow to turbulent flows in high speed mixers. The principles and practice of LDV are discussed in detail by Durst, Melling, and Whitelaw (1981) and have been recently reviewed by Adrian (1996). Here we provide a brief description of the principle of operation, and review its application to non-Newtonian flow problems.

In LDV, the Doppler frequency shift of laser light that has been scattered by small particles (typically 0.1 to 10 μm in diameter) moving with the fluid is measured. The frequency of light from a laser scattered by a particle moving relative to the laser is changed by an amount – the Doppler shift ν – that depends only on the velocity of the particle and the scattering geometry. The use of a laser as the source is critical so that the incident light is monochromatic and coherent. For incident light in the visible portion of the spectrum and velocities on the order of a cm/s, this frequency shift is on the order of 10^4 Hz. Since this shift is extremely small compared to the frequency of laser light (of order 10^{14} Hz), an optical heterodyne technique is used: the scattered light is mixed or heterodyned with another beam at nearly the same frequency. This mixing or superposition of two waves gives the frequency difference between the two beams directly. If the scattered light from two beams focused at the same point in the flow (through different angles) is mixed, the techique is referred to as dual beam scattering (Fig. 2). Only the component of velocity normal to the bisector of the beams and in the same plane is measured, and hence the frequency shift measured is independent of the position of the detector. The frequency shift is given by

$$\nu = \underline{v} \cdot (\underline{s}_2 - \underline{s}_1)/\lambda \qquad (1)$$

where $\underline{v}$ is the velocity, $\underline{s}_1$ and $\underline{s}_2$ are the propagation directions of the two beams, and λ is the wavelength of light. Thus the velocity is directly proportional to the Doppler shift ν of the scattered light.



$$\nu_1 = \nu_o + \underline{v} \cdot (\underline{r} - \underline{s}_1) / \lambda$$
$$\nu_2 = \nu_o + \underline{v} \cdot (\underline{r} - \underline{s}_2) / \lambda$$
$$\nu = \underline{v} \cdot (\underline{s}_2 - \underline{s}_1) / \lambda$$

Fig. 2. The dual beam scattering configuration for laser Doppler velocimetry. Scattered light from two incident beams focused at the same point in the flow (through different angles) is mixed at the detector. The resulting Doppler shift ν is independent of the position of the photodetector.

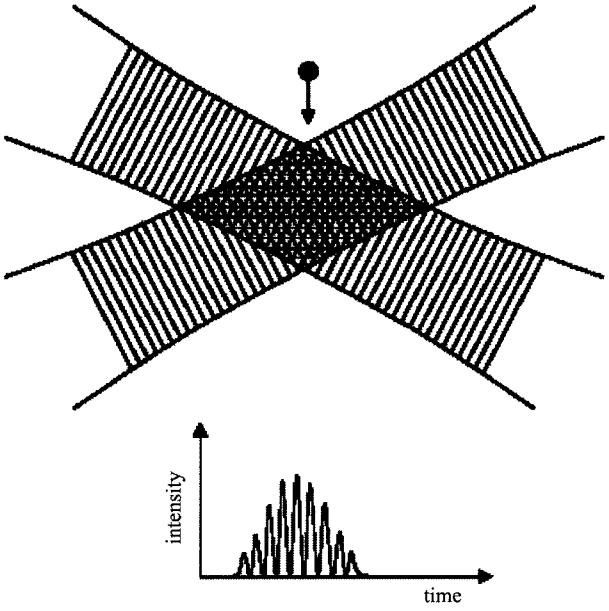


Fig. 3. The intersection of the two beams in laser Doppler velocimetry defines a set of interference fringes (oriented horizontally in the figure). A particle passing through the measuring volume scatters light as it passes through this set of bright and dark fringes; so the measured intensity fluctuates in time. The spacing of the intensity peaks is proportional to the known fringe spacing and the particle's velocity.

A useful heuristic device for visualizing the method is the fringe model, suggested by Fig. 3. The intersection of the two beams in the flow defines the measuring volume. In that measuring volume, the wavefronts are quasi-planar, and the interference of these wavefronts creates a pattern of parallel bright and dark interference fringes. A particle moving with the fluid through these interference fringes scatters light; the intensity of the scattered light oscillates at a frequency that is related only to the spacing between the fringes and the component of the particle's velocity normal to the fringes. The scattered light is collected by a photomultiplier, and can be processed to recover the Doppler frequency, and hence the velocity of the particle, using a variety of digital signal processing methods.

The size of the measuring volume defines the spatial resolution of the technique. Typical dimensions range from 0.1 mm × 0.8 mm × 0.1 mm; with special design of the optical system, these values may be reduced by more than an order of magnitude (Adrian, 1996). The time resolution is essentially related to the signal processors frequency response. For a range of processors, the measurement time is of the order of the time for a single Doppler burst (that is, for a particle to pass through the measuring volume), which is normally short in comparison to the flow time scale. As a result, these processors are capable of resolving the velocity as a function of time.

LDV has been particularly valuable in identifying the onset of instabilities in viscoelastic flows. Contraction flows have received perhaps the most attention (Kramer and Meissner, 1980; Lawler *et al.*, 1986, Raiford *et al.*, 1989; Quinzani *et al.*, 1994, 1995; McKinley *et al.*, 1991; Castro and Pinho, 1995; Subramanian and Picot, 1994; Naka *et al.*, 1999; Munstedt *et al.*, 2001). There was early success in the axisymmetric contraction problem in identifying a transition from a steady axisymmetric flow to a time-dependent, three dimensional flow (Lawler *et al.*, 1986). The high spatial resolution of LDV was critical in this instance: because the transition occurred locally near the entrance to the contraction, it had not been readily apparent in earlier, large scale, global flow visualization experiments. Subsequent studies revealed the effects of fluid rheology, inertia, and contraction ratio on the transitions, and the rich sequence of transitions that occur with increasing elasticity (Raiford *et al.*, 1989; McKinley *et al.*, 1991). This work had a significant impact on the use of this problem as a benchmark for numerical simulations.

LDV has been used to study elastic instabilities and transitions in a number of other flow problems. Critical conditions, critical frequencies, and the spatio-temporal symmetry of elastic instabilities in Taylor-Couette flow (Muller *et al.*, 1993; Groisman and Steinberg, 1998), Taylor-Dean flow (Joo and Shaqfeh, 1994), and flow past a confined cylinder (McKinley *et al.*, 1993) have been measured. The fast temporal response of LDV makes it an ideal tool for studies of the flow spectrum. High resolution power spectra of the time dependent velocity at a point in the flow often reveal transitions that are undetectable through direct examination of global flow visualizations. In these problems, as in the axisymmetric contraction problem, higher order transitions were first made evident through the appearance of new characteristic frequencies in the power spectrum. This is illustrated by Fig. 4, taken from early work on the 4:1 axisymmetric contraction problem (Lawler *et al.*, 1986).

LDV has been applied to a number of other classic flow problems for viscoelastic fluids. These include viscoelastic flow past a sphere (Bisgaard, 1983; Arigo *et al.*, 1995; Arigo and McKinley, 1998), between eccentric cylinders (Lawler *et al.*, 1986; Berker *et al.*, 1995; Dris and Shaqfeh, 1998), lid-driven cavity flow (Pakdel *et al.*, 1997; Pakdel and McKinley, 1998), viscoelastic flow past a periodic array of cylinders (Chmielewski and Jayaraman, 1993), flow in a corrugated channel (Yalamanchili *et al.*, 1993, 1995; Koshiba *et al.*, 1999), and in a constant extensional rate channel (Shirakashi *et al.*, 1998).

In many of these flows, steady velocity profiles have been directly compared to numerical simulations of the

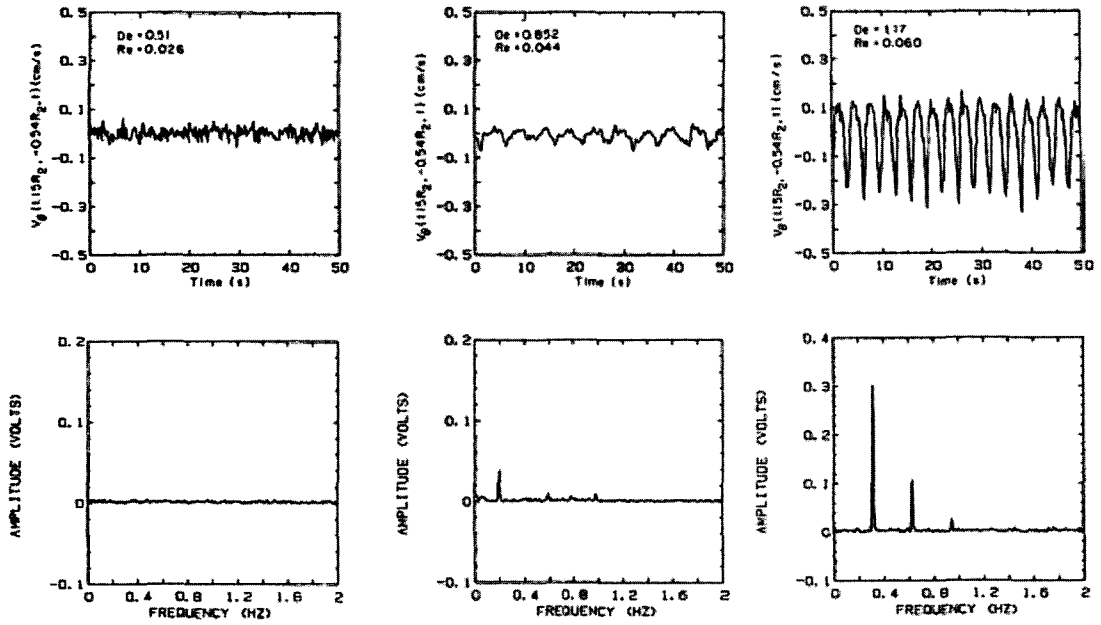


Fig. 4. LDV measurements of the azimuthal velocity v_θ at a point just upstream of the lip of a 4:1 axisymmetric sudden contraction. The top row of figures show $v_\theta(t)$ at increasing elasticity, parameterized by the Deborah number De. The bottom row of figures are the corresponding frequency spectra, illustrating the emergence of time-periodic flow states. (Reproduced from Lawler *et al.*, 1986).

flow as tests of constitutive models. The potential of LDV for high spatial resolution and high accuracy over a broad dynamic range make it well-suited for such comparisons; however, the pointwise nature of the technique often makes reconstruction of the full velocity field intractable even in steady flows. The spatial resolution has also been exploited to examine wall slip of a polymer melt in an extrusion die (Piau *et al.*, 1995). In a few cases, more complex, processing flows of non-Newtonian fluids, including flow in a twin-screw extruder (Chiruvella *et al.*, 1996) and flow in a disc turbine (Mishra *et al.*, 1998) have been studied.

The excellent temporal response of LDV systems has been exploited by a number of authors interested in the use of polymers as drag-reducing agents in high speed or turbulent flows. Turbulent pipe flow of polymer solutions has received considerable attention (Hoyer and Gyr, 1996; DenToonder *et al.*, 1997; Ptasinski *et al.*, 2001; Savvas *et al.*, 1994). The effects of polymer solutions on the interaction between a free surface and a polymer solution jet (Tanaka *et al.*, 2000), steady streaming flow induced by an oscillating cylinder (Vlassopoulos and Schowalter, 1993), and in a free-mixing shear layer (Scharf, 1994) have also been examined.

LDV has been successfully combined in a few studies (Baaijens *et al.*, 1994, 1995; Quinzani *et al.*, 1994, 1995; Bedford and Burghardt, 1996; Davidson *et al.*, 1993a, b) with flow induced birefringence measurements to provide information about both the velocity at a point and the integrated stresses. Flow induced birefringence integrates across the flow field, so these studies have been confined to two dimensional geometries, including planar contraction flow, flow past a cylinder in a channel, and a periodically constricted channel. The combined stress and velocity data have allowed for detailed, quantitative comparisons to numerical simulations and testing of constitutive equations.

Since the laser Doppler technique measures light scattered by particles in the flow, the fluid itself must be transparent to the wavelength of the illuminating laser. This is a severe constraint and precludes its use in non-Newtonian fluids that are opaque or contain many small particles of a different refractive index than the fluid. (Although, through careful refractive index matching of the solid and liquid phases, Leal and co-workers (Lyon and Leal, 1998, Koh *et al.*, 1994) have used LDV to study concentrated suspensions in two dimensional channel flow.) The optics and signal processing equipment make LDV a fairly expensive technique to implement. Alignment of the optics can be time-consuming and challenging, particularly for simultaneous measurement of multiple components of the velocity, which requires that multiple pairs of beams be focussed at the same point in the flow. In addition, LDV yields only pointwise information about the velocity field, making it difficult or impossible to reconstruct the global flow field. For these reasons, it has been used by a relatively small number of researchers in non-Newtonian fluid mechanics.

2.2. Homodyne light scattering spectroscopy

While LDV measurements can in principle be differentiated to provide velocity gradient information, there are a number of inherent limitations to obtaining the gradient of the velocity in this way. There are difficulties in differentiating experimental data, and in situations where there are sharp gradients in velocity the problem is compounded by the averaging over the measuring volume that occurs in LDV. Thus, in small scale flow experiments or in flows with steep boundary layers where spatial resolution of the velocity field is an issue, it is desirable to measure the velocity gradient directly.

If the scattered light from a particle in a flowing liquid is mixed with itself, rather than with the incident light or light scattered by a beam propagating in a different direction as in LDV, the resultant frequency spectrum becomes insensitive to the mean velocity. Under certain conditions, this homodyne light scattering may be used to determine the velocity gradient at the measuring volume. Homodyne light scattering spectroscopy for the measurement of velocity gradients in steady, laminar flows has been presented in detail by Fuller and co-workers (Fuller *et al.*, 1980; Fuller, 1995). Its use has been extended to transient but repeatable flows by Wang *et al.*, (1994).

Briefly, for the scattering geometry shown in figure 1, the incident and scattered wave vectors are $\underline{k}_i$ and $\underline{k}_s$, respectively, and the scattering vector $\underline{q}$ is equal to $\underline{k}_i - \underline{k}_s$. The magnitude of the scattering vector $\underline{q}$ is $q = (4\pi n_0/\lambda_0)\sin(\theta/2)$, where n_0 and λ_0 are the refractive index of the solvent and the wavelength of light in a vacuum, respectively. Thus, we have a length scale 1/q and a characteristic length L associated with the width of the incident beam. The diffusion time scale is $(Dq^2)^{-1}$, where D is the particle diffusion coeffient. The time scale associated with the velocity gradient is $(q\gamma L)^{-1}$, where γ is the magnitude of the velocity gradient. The homodyne correlation function will be dominated by the time scale associated with the velocity gradient as long as it is much smaller than the time scale for diffusion. Hence, as we outline below, measurement of the homodyne correlation function can give a direct measurement of the velocity gradient γ; in addition, the technique can provide information about the flow type, or the strength of the flow.

For a Gaussian intensity profile of the incident beam, and a linear flow field of the form $\underline{v} = \underline{\underline{\gamma}} \bullet \underline{x}$ where $\underline{v}$ is the local fluid velocity and $\underline{x}$ is the position, with

$$\underline{\underline{\gamma}} = \begin{bmatrix} 0 & \gamma & 0 \\ \alpha\gamma & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix} \tag{2}$$

where $0 < \alpha < 1$ is the flow strength parameter, the homodyne correlation function $F_2(q, t)$ is given by (Wang *et al.*, 1994):

$$F_2(q,t) = \beta\exp\left\{-\frac{1}{2}q^2\gamma^2L^2t^2\cos^2\frac{\theta}{2} \times \left[\left(\frac{1+\alpha}{2}\right)^2\sin^2 2\psi + (\cos^2\psi - \alpha\sin^2\psi)^2\right]\right\}. \tag{3}$$

Here β is an experimental constant that depends on the detector optics and alignment, t is time, and ψ is the angle between the scattering vector $\underline{q}$ and the flow direction. If the flow direction and the scattering vector are coincident ($\psi = 0°$), $F_2(q, t)$ reduces to the following:

$$F_2(q,t) = \beta\exp\left\{-\frac{1}{2}q^2\gamma^2L^2t^2\cos^2\frac{\theta}{2}\right\} = \beta\exp\{-[\Gamma(0^\circ)]^2t^2\} \tag{4}$$

The decay constant $\Gamma(0°)$ is proportional to $q\gamma L$, thus a measurement of the homodyne correlation function at $\psi = 0°$ yields a value of the local velocity gradient γ. When $\psi = 90°$, we obtain

$$F_2(q,t) = \beta\exp\left\{-\frac{1}{2}q^2\gamma^2L^2t^2\alpha^2\cos^2\frac{\theta}{2}\right\} = \beta\exp\{-[\Gamma(90^\circ)]^2t^2\} \tag{5}$$

The decay constant $\Gamma(90°)$. $\sim q\,\gamma\alpha L$. Thus, a second measurement of the homodyne correlation function at $\psi = 90°$ gives the flow strength parameter α. For absolute determination of the velocity gradient, however, the characteristic beam width L must also be known.

Fuller and co-workers (1980) verified this technique using Newtonian flow in a Couette device and flow due to a rotating cylinder in a large bath of fluid; they then examined flow in a four-roll mill. Fuller and Leal (1980) considered the flow of a dilute polymer solution in a range of flows generated in the four roll mill, and used homodyne light scattering along with flow birefringence. Wang *et al.* (1994) have demonstrated the extension of homodyne light scattering techniques to repeatable time-dependent flows generated in a two roll mill and reported a time resolution of 0.05 s.

The technical challenges associated with homodyne light scattering spectroscopy are more severe than in laser Doppler experiments and are comparable to those in conventional dynamic light scattering. The analysis requires that scatterers meet the Rayleigh-Debye condition (Berne and Pecora, 1976; Fuller *et al.*, 1980); as a result fluids must be carefully filtered to remove large particles and surfaces of the flow apparatus must be scrupulously cleaned to remove dust and any potential for stray scattering. Like LDV, it provides a pointwise measurement; to determine velocity gradients across the whole flow field, the scattering volume must be spatially translated with respect to the flow field. In addition, the measurement requires that the flow direction be aligned with the scattering vector; hence in complex flows one must first ascertain the flow direction. As a result, use of homodyne light scattering to measure veloc-

ity gradients in non-Newtonian fluids has been limited to a very small number of flows. These include Taylor-Couette flow (Tsunashima, 1995), and flows generated in two and four roll mills (Ng and Leal, 1993; Yavich *et al.*, 1998; Lee and Muller, 1999; Wang *et al.*, 1994).

3. Marker displacement techniques

A range of related velocity measurement techniques rely on directly observing small, marked regions of fluid and recording the positions of the markers at two or more times. Markers may be small particles, bubbles, or molecules that are photo-activated to produce a change in color, fluorescence, or phosphorescence. In the conventional implementation of these marker displacement techniques, global velocity fields in two dimensional domains are measured by illuminating a plane with a light sheet, and capturing images of the positions of the markers on film or with a CCD camera at multiple times. Marker (and thus fluid) velocity is determined from the displacement of the markers during the known time between exposures. The marker velocity is simply:

$$\underline{v}(\underline{x}, t) = \Delta\underline{x}/\Delta t \tag{6}$$

where $\Delta\underline{x}$ is the displacement of the particle in the plane and Δt is the time between exposures. Techniques that use particles as markers, especially particle image velocimetry, have been discussed in a thorough review by Adrian (1991). Techniques that rely on molecular markers have been discussed by Miles and Lempert (1997) and Koochesfahani and co-workers (1997). A key advantage of these techniques over LDV and homodyne light scattering is their ability to provide quantitative measurement of whole, instantaneous velocity fields. Here we will discuss two marker displacement techniques, particle image velocimetry and molecular tagging velocimetry.

3.1. Particle image velocimetry

The use of particles as markers, and the use of particle motion to provide information about flow fields, has a rich history in fluid mechanics. Flow visualization has long been used to provide qualitative information about velocity fields, and many beautiful examples of flow visualization in non-Newtonian flows can be found in Boger and Walters (1993). Here, we discuss extensions of these techniques over the past several years that allow quantitative velocity vector fields to be recovered. The basic set up is shown in Fig. 5.

A sheet of laser light, typically about 1 mm in thickness, is used to illuminate the measurement plane. Scattering from small particles, typically in the size range of 1 to 20 μm, moving with the fluid is imaged onto a camera located at 90° to the sheet. Images of the measurement region are captured either on film or with a CCD camera, and transferred to a computer for automated analysis. The entire image field is divided into subsections or interrogation regions, and analyzed subsection by subsection.

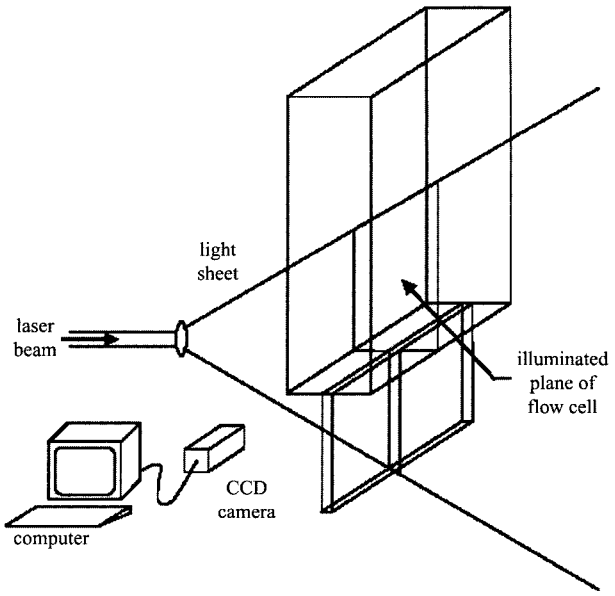


Fig. 5. A schematic of a particle image velocimetry (PIV) system. A cylindrical or Powell lens converts the incident laser beam into a sheet of laser light, which illuminates a plane in the flow cell. A CCD camera is placed at 90° to the light sheet, and captures images of particles moving in the flow. Images are transferred to a computer for subsequent image processing.

At very low particle concentrations, velocities can be determined by particle tracking velocimetry. Particle images are sparse enough that the center of each particle can be located and individual particles can be tracked to determine the displacement of each particle between exposures. Due to the low particle concentration, there is little ambiguity in associating images of particles between exposures or due to crossing particle trajectories, so analysis of the exposures is straightforward. However, since the velocity field is sampled only at the random locations of the particles, data is obtained at random, often widely spaced points; i.e., many subsections of the image field may contain no particles.

Spatial resolution can be improved by increasing the particle concentration. When particles are present at very high concentrations, the images of the particles overlap in the image plane, and random phase differences between the images of particles create interference or speckle patterns. Under these conditions, the technique is referred to as Laser Speckle Velocimetry, and the fluid velocity is measured by determining the speckle displacement. It is frequently difficult to seed the flow at concentrations sufficiently high to produce useful speckle fields. At inter-

mediate concentrations, however, the particle concentration is high enough that each subsection contains many particle images, but not enough to produce a speckle image. The large number of particle images makes tracking individual particles difficult, but statistical approaches allow one to infer the average displacement of groups of particles in each subsection or interrogation region. This mode of velocimetry is referred to as particle image velocimetry (PIV).

Typically, CCD images are captured to computer. The displacement in each subsection is then calculated by locating the peak of the cross correlation of the intensity fields of corresponding subsections of a pair of exposures. This robustly recovers the mean displacement provided the variation in displacement from particle to particle in the subsection is small. This calculation is repeated for each subsection to recover the velocity field across the entire image field (Adrian, 1991; Willert and Gharib, 1991). A number of extensions have been developed to improve spatial resolution, increase the dynamic range of velocity resolution, reduce discretization and aliasing errors, and improve outlier rejection (Westerweel, 1993; Adrian, 1997). The accuracy of the techique has been shown to be 1% in velocity and 4% in its derivatives (strain, vorticity) for very smooth, planar flows. The maximum and minimum velocities that can be measured are determined by the time separation Δt between images. Pulsed lasers can be used to both shorten the duration of an exposure and to decrease the time separation between exposures; frequency doubled pulsed Nd:Yag lasers allow 10 ns pulses at rates up to 50 Hz, and two of these lasers can be combined to produce virtually any separation between pulses.

One of the earliest applications of PIV to viscoelastic fluids, by Pakdel and McKinley (1997), demonstrated the potential of PIV by examining steady flow in circular pipes, flow in a lid-driven cavity, and time-dependent free-surface extensional flow in a liquid filament. Pakdel and co-workers (1998; 1997a, b) combined PIV with LDV and flow visualization to probe the spatial and temporal characteristics of secondary flows in the lid-driven cavity problem as a function of cavity aspect ratio and fluid elasticity. PIV has been used by Shiang *et al.* (1997; 2000) to map elastic transitions in viscoelastic flow past a confined circular cylinder. Detailed PIV measurements have been made in the classic problem of viscoelastic flow past a sphere (Fabris *et al.*, 1999; Arigo and McKinley, 1998), in the swirling flow in a torsionally driven cylindrical cavity (Stokes *et al.*, 2001a, b), and in an axisymmetric contraction-expansion (Rothstein and McKinley, 1999; 2001). PIV has also been used in a few studies of non-Newtonian fluids at high Reynolds numbers to examine drag reduction effects (Arora *et al.*, 2002; van Doorn *et al.*, 1999; Saadeh *et al.*, 1997).

PIV is most easily applied to 2D flows in which the particles remain in the illumination plane. Recently, Baumert and co-workers (1997) described a novel implementation of PIV in flows with nearly closed pathlines where the primary flow is orthogonal to the plane of illumination. The r-z plane of a Taylor-Couette (concentric cylinder) apparatus was illuminated and extremely low levels of particle seeding and long exposure times were used. The primary flow is in the azimuthal direction and, for weak secondary flows, in-plane particle displacements are small. However, by allowing each particle to pass through the plane multiple times during an exposure, and performing the cross correlation on pairs of these long exposures, secondary flows that were four to six orders of magnitude weaker than the base flow could be resolved. An example of an image pair and the resulting (r,z)-velocity vector map is shown in Fig. 6 for a weak, viscoelastic disturbance flow in the Taylor-Couette geometry. Another development in PIV that may be increasingly important is its use in microfluidic devices (Santiago *et al.*, 1998). Sample data from flow of a polymer solution through a microfluidic contraction are shown as Fig. 7 (Chen, 2001). Microfluidic platforms offer unique opportunities for measurements of both velocity fields, using PIV through a fluorescence microscope, and conformation fields, using fluorescently labeled DNA as a model polymer (Perkins *et al.*, 1997, Shrewsbury *et al.*, 2001).

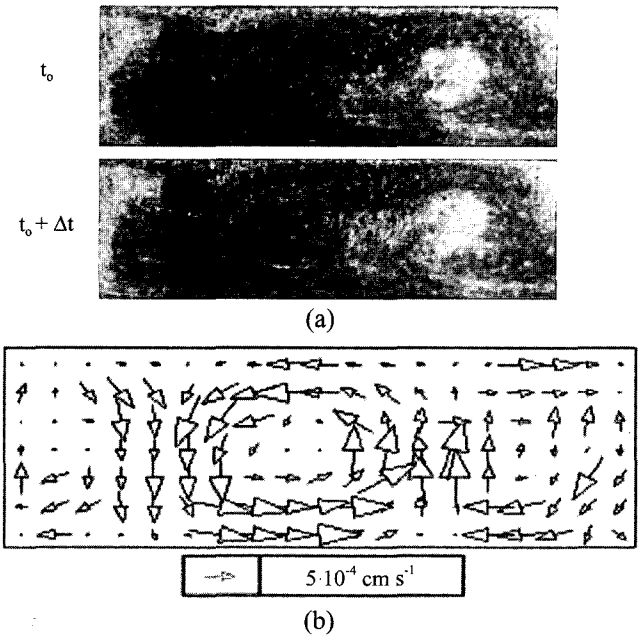
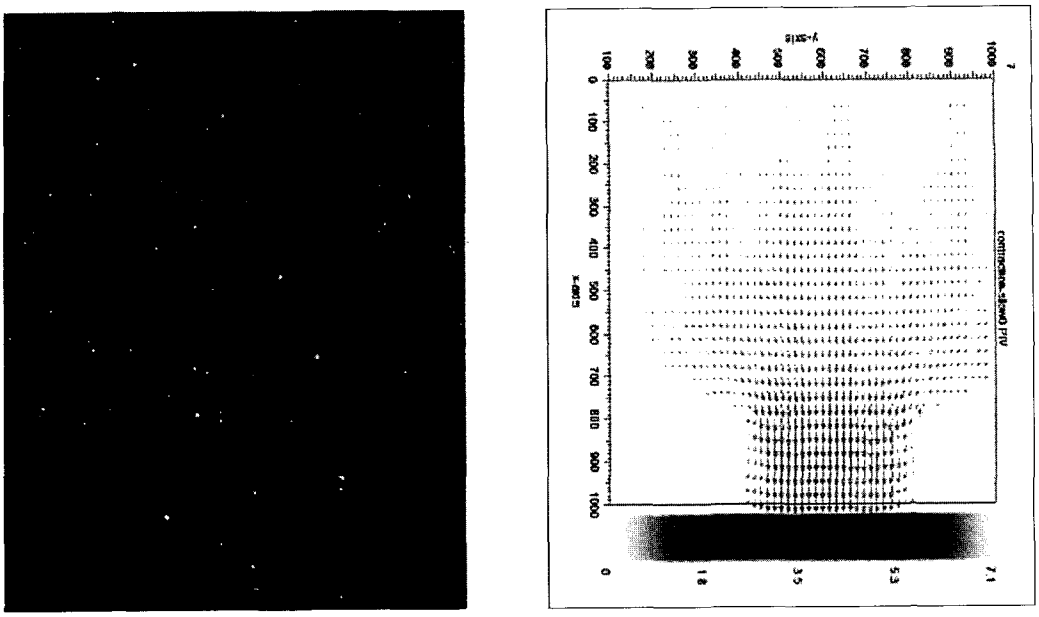


Fig. 6. A PIV image pair (a) and resultant velocity vector map (b). A small, central section of the r-z plane of a Taylor-Couette cell was imaged; the inner cylinder is at the bottom of each image, the outer cylinder is at the top, and the primary flow is out of the plane of the image. (Reproduced from Baumert *et al.*, 1997).



(a) (b)

Fig. 7. A sample PIV image (a) and velocity vector map (b) from flow in a microfluidic device. The downstream channel is 300 μm across. The channel depth normal to the image plane is 60 μm. PIV was performed through a fluorescence microscope, using fluorescent microspheres as markers.

Owing to the simple set up – a laser, a cylindrical or Powell lens to generate the light sheet, camera and computer are the only equipment required – PIV is relatively inexpensive and easy to undertake. As long as the fluid is optically transparent, and particles track the flow and can be homogeneously dispersed in it, PIV is capable of producing accurate, high quality, instantaneous measurements of velocity fields in 2-D flows, and is becoming increasingly popular for investigating non-Newtonian flows. Several extensions of PIV to fully threedimensional flows have been described for Newtonian fluids (Lawson and Wu, 1999; Fabry, 1998; Fincham and Spedding, 1997; Halloin and Jottrand, 1994), and the routine application of 3D PIV to non-Newtonian fluids in the near future is likely.

3.2. Molecular tagging velocimetry

The use of even small particles as markers can sometimes present problems. Uniform dispersal of the particles may be difficult, the particles may not follow the flow, and the high levels of particles necessary in PIV can lead to secondary scattering problems. In molecular tagging velocimetry (MTV), in place of micron size particles, certain types of photoactive molecules are used as flow markers. The liquid is seeded with the photoactive molecules and a laser is used to "tag" or excite the molecules in a grid or pattern. The imaging of this grid at successive times (within the lifetime of the "tag") allows determination of displacement in much the same way as in PIV. These molecular tagging methods have been demonstrated with a range of compounds, including photochromic molecules, caged fluorescent dye molecules, and phosphorescent supramolecules. In all these methods, the lifetime of the "tagged" molecular marker is a key parameter: it must be long enough relative to the flow time scale to allow imaging after sufficient displacement. In addition, the "tagging" process must be rapid (Miles and Lempert, 1997; Koochesfahani *et al.*, 1997).

When photochromic dyes are used, the technique is sometimes referred to as laser induced photochemical anemometry (LIPA). A UV laser is typically used to induce a color change in a photochromic dye and a white light source is used to interrogate the displaced regions of dye. The tagging process takes place in a few nanoseconds, the dyes can have lifetimes of up to a few minutes, and the process is completely reversible and thus reusable. When a caged fluorescent dye such as fluorescein is used, the method has been referred to as "photo-activated non-intrusive tracking of molecular motion" (PHANTOMM). Here, a fluorescein molecule is chemically modified to render it non-fluorescent. The "caging" group is removed on absorption of UV light, releasing the fluorescent dye, which can be interrogated through its luminescence on excitation with the appropriate wavelength radiation. Two wavelengths of light are thus needed – one to photolytically cleave the cage and one to excite the fluorescence. The kinetics of the cage-breaking reaction are important; the reaction must be fast relative to flow time scales. In addition, the reaction is not reversible. A schematic of this method is shown in Fig.

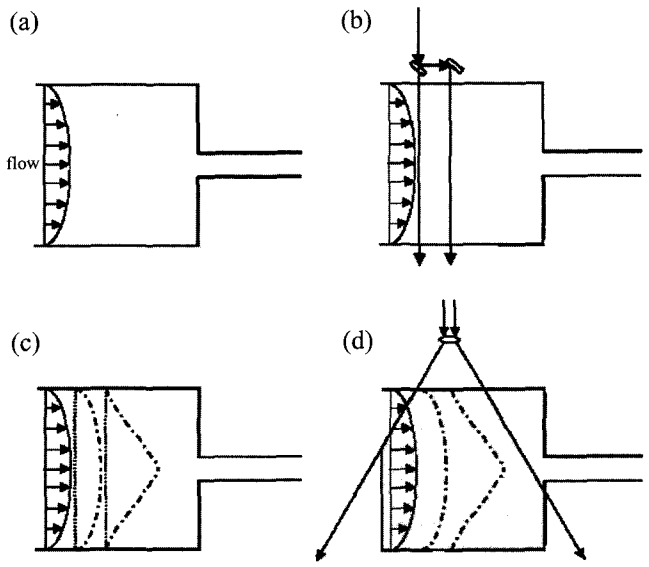


Fig. 8. A schematic of an implementation of molecular tagging velocimetry (MTV). (a) The flow is seeded with a caged fluorescent dye molecule. (b) A UV laser beam is used to remove the caging group and release the fluorescent dye along a pattern of lines. (c) The dye, released in the positions indicated by the dotted lines, is convected by the flow to positions indicated by the dashed lines. (d) The positions of the dye molecules after convection is imaged by exciting the fluorescent molecules with a laser at the appropriate wavelength.

8. Phosphorescent compounds may also be used for MTV; excitation of the compound by light is used to produce an excited state that is interrogated through its phosphorescence emission as it returns to the ground state (Koochesfahani *et al.*, 1997).

In addition to eliminating problems associated with introducing particles to the flow, molecular tagging velocimetry offers some advantages in terms of measuring flows with out-of-plane motion and extensions to three-dimensional measurements. Since only the tagged molecules contribute to the measurement, the motion of molecules outside of the tagged region into or out of the interrogation plane does not contribute to the spatial correlation of the markers during the processing of the images. While MTV has been demonstrated on a number of Newtonian flows (Koochesfahani *et al.*, 1997; Miles and Lempert, 1997; Lempert and Harris, 2000; Chu *et al.*, 1993), it has yet to be utilized in non-Newtonian flows although it is well-suited to use in these systems.

4. Nuclear magnetic resonance imaging

The methods described above all require that the fluid be optically transparent. When the fluid is opaque, velocity measurements by nuclear magnetic resonance imaging (NMRI) techniques are possible. The measurement of flow using NMR was first reported in the 1950's; early uses of NMRI were largely associated with medical applications. Recently, NMRI has been applied to a number of non-Newtonian systems of fundamental and industrial importance. NMR methods have a number of strengths, including noninvasiveness, the ability to work in optically opaque systems, and the ability to work in systems that are reactive, abrasive, or need to be isolated. Since NMR can distinguish between different states of matter and different chemical species, it is possible to obtain flow information for each chemical species in the system. The technique offers the potential for 3-dimensional imaging, and image projections in any direction are possible. The potential also exists for measuring higher order parameters, including acceleration and fluctuations of velocity. Spatial resolutions of 10-40 microns have been reported; because of sensitivity limitations, however, temporal signal averaging is required and NMRI has been used primarily in steady or slowly varying flows. NMR of fluid motion is discussed in detail by Callaghan (1991) and has been recently reviewed by Gladden (1994), Gladden and Alexander (1996), Fukushima (1999), and Callaghan (1999).

Briefly, NMR spectroscopy detects nuclear spins precessing in a strong magnetic field. In an applied static magnetic field B_o, a sample of interest containing protons will develop a bulk magnetization parallel to the applied field that is proportional to the local spin density. By applying a radio frequency (rf) magnetic field pulse at the appropriate frequency ω_0, (the Larmor frequency) where

$$\omega_0 = \gamma_M B_o \tag{7}$$

and γ_M is a constant for each species of nuclei, the magnetization is rotated 90° away from the static field. The time-dependent signal due to the precession of the transverse magnetization around the local magnetic field at the local Larmor frequency is measured. To observe magnetic resonance phenomena as a function of spatial position, a magnetic field gradient must be applied – under these conditions the Larmor frequency depends on the position of the spin. By detecting the signal and measuring its frequency, one can measure the position of a region of spin. To determine spatial position in 3 dimensions, linear gradients must be applied and varied in x, y, and z directions.

One of a number of methods for using NMR for flow imaging involves using an rf pulse sequence to selectively "tag" the sample in a selected slice or pattern. At a later time, the 3D distribution of these tagged spins is interrogated, and from the displacement of the tags the velocity (and concentration) profiles can be obtained. This method has similarities to the molecular tagging velocimetry method suggested by figure 8; here the tag is nuclear spin. Tracking the phase evolution of the transverse spin magnetization is an alternate method that provides higher spa-

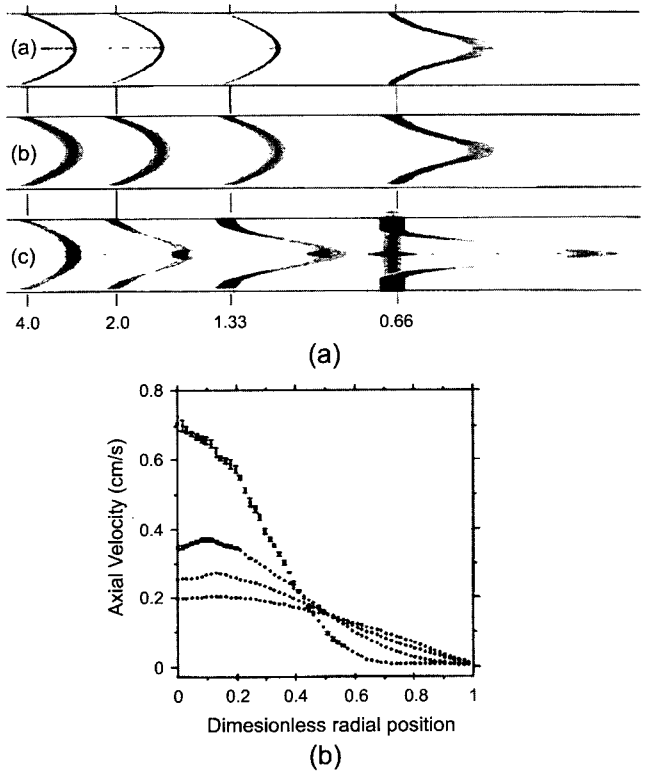


Fig. 9. (a) Time-of-flight NMR flow imaging of flow in a 3:1 axisymmetric sudden contraction. Displacement profiles are shown for glycerin (top), an isotropic 35 wt% HPC solution in water (middle), and a liquid crystalline 50 wt% HPC solution in water (bottom). (b) Quantitative NMR imaging of the axial velocity of the liquid crystalline HPC solution upstream of the contraction plane. The profiles correspond to axial locations equal to 0.66, 1.3, 2, and 3 upstream tube radii before the contraction plane. (Reproduced from Gentzler *et al.*, 2000).

tial resolution of the velocity field. Velocities have been measured from a few tens of microns per second to of order 100 cm/s.

Because of its versatility and unique ability to probe optically opaque systems, NMRI has been widely used in non-Newtonian systems. Several reviews of NMRI have recently appeared which include summaries of the rich literature on NMRI applications to complex fluids (Gladden, 1994; Gladden and Alexander, 1996; Fukushima, 1999; Callaghan, 1999).

Work on polymer solutions and melts, wormlike micelles, liquid crystalline polymers, foods, colloids, and granular materials has been particularly well covered by Callaghan (1999), and the interested reader is referred to that work and the references therein. Here, we show a single illustration of NMRI, applied to a liquid crystalline solution of hydroxypropylcellulose (HPC) in water in flow through a 3:1 axisymmetric abrupt contraction (Fig. 9) to suggest the qualitative and quantitative information that can be obtained (Gentzler *et al.*, 2000).

The experimental requirements for NMRI are non-trivial. An NMR spectrometer is expensive and the need for a high magnetic field translates into a relatively small working space: a typical magnet may have a working space on the order of 2 to 10 cm across. Callaghan (1999) has described a number of "rheo-NMR" geometries – including cylindrical Couette cells, cone and plate cells, and four-roll mills – which will fit inside the vertical bore of a NMR superconducting magnet; and applications in more complex geometries are rapidly appearing in the literature. Moreover, the versatility of NMR imaging methods, and their potential for providing complementary information over a range of length and time scales (concentration, molecular orientation, nematic director alignment, shear induced mesophase reordering) provide unique opportunities for the study of non-Newtonian materials.

5. Summary

The number of tools available for obtaining detailed, quantitative velocity fields in non-Newtonian fluids has grown considerably in recent years. Advances in laser technology, image capture, computer-aided image processing, and NMR methods have increased the versatility and dynamic range of particle imaging and NMR imaging techniques, and we expect to see further expansion of their use in complex flows relevant to rheology and materials processing. In optically transparent fluids, the full spectrum of techniques described in this review is available. LDV will provide highly spatially and temporally resolved pointwise measurements of velocity, but is relatively expensive to implement. Homodyne light scattering spectroscopy can provide direct, pointwise velocity gradient data provided the flow is repeatable, but requires independent information about the flow direction and is technically challenging to implement. PIV is emerging as an extremely valuable and versatile tool for obtaining global 2D velocity fields in flows which can be seeded with small particles. It is relatively easy and inexpensive to implement unless very high speed or rapidly changing flows must be measured; in these cases, the costs of pulsed lasers and high speed cameras must be considered. NMR imaging, which is the only technique that can be used in optically opaque liquids, can provide 3D velocity maps as well as complementary data about concentration and mesoscopic structure. The spatial constraints imposed by the requirement of generating high magnetic fields in the sample and the expense of a NMR spectrometer at present impose some limitations on its use.

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