Available online at www.sciencedirect.com
ScienceDirect
Ultrasonics 44 (2006) e477–e482
Ultrasonics
www.elsevier.com/locate/ultras

Inline ultrasonic rheometry by pulsed Doppler

David M. Pfund *, Margaret S. Greenwood,
Judith A. Bamberger, Richard A. Pappas

Pacific Northwest National Laboratory, Battelle Boulevard, MS K7-15, Richland, WA 99352, USA

Available online 2 June 2006

Abstract

This will be a discussion of the non-invasive determination of the viscosity of a non-Newtonian fluid in laminar pipe flow over the range of shear rates present in the pipe. The procedure used requires knowledge of the flow profile in and the pressure drop along a long straight run of pipe. The profile is determined by using a pulsed ultrasonic Doppler velocimeter. This approach is ideal for making non-invasive, real-time measurements for monitoring and control.

Rheograms of a shear thinning gel will be presented. The operating parameters and limitations of the Doppler-based instrument will be discussed. The most significant limitation is velocity gradient broadening of the Doppler spectra near the walls of the pipe. This limitation can be significant for strongly shear thinning fluids (depending also on the ratio of beam to pipe diameter and the transducer's insertion angle).

1. Introduction

Ultrasonic methods are well-established performers in a myriad of industrial applications and have been receiving increased attention as chemometric tools in process analytical chemistry [1–3]. Ultrasonic velocity, attenuation, reflection coefficients, and scattering amplitudes are measurable parameters related to fundamental physical properties of fluids and slurries of interest to food processors and manufacturers of consumer products [4–6]. Ultrasonic methods can also provide flow-rate and rheological information [7,8] on the contents of a process stream that can be invaluable in the monitoring and control of product quality. There are commercial devices for monitoring average flow rates based on transit-time measurements [9]. One commercial device (from Met-Flow, SA) provides velocity profiles generated with range-gated Doppler measurements [10]. The viscosity of a process stream is potentially an indicator of its composition, solids loading, and extent of mixing. Viscosity could be a useful control variable that indicates the need for heating or dilution. Traditional off-line measurements are slow and samples can be difficult to extract. Extracting samples can slow down production and so require increased capacity in the whole process. An alternative viscosity measurement that does not require sampling, and that is fast would be more useful than traditional techniques.

The concept of non-invasive inline rheometry has been developed for both nuclear magnetic resonance [11] and ultrasonic measurement methodologies. The ultrasonic method, ultrasonic Doppler velocimeter (UDV), is the more practical and economical approach for most manufacturing applications. We have developed an ultrasonic Doppler velocimetry technique [12] that provides non-disruptive rheological measurements and has been successfully demonstrated on a pseudoplastic fluid flowing at a nominal maximum flow rate of $33\,\mathrm{l\,min^{-1}}$ in a 2-in. (0.05 m) diameter pipe. The design has recently been refined to address limitations in the maximum measurable flow rate, the velocity profile accuracy, and the Doppler signal-to-noise ratio that have tempered the application of the inline ultrasonic rheometer technology to industrial monitoring needs.

* Corresponding author. Tel.: +1 509 375 3879; fax: +1 509 375 3865.
E-mail address: david.pfund@pnl.gov (D.M. Pfund).

0041-624X/$ - see front matter © 2006 Elsevier B.V. All rights reserved.
doi:10.1016/j.ultras.2006.05.027

In this work, rheological measurements of non-Newtonian fluids were made non-invasively with the UDV-based technique. A viscometric flow was created within the transport pipeline itself and shear stress and shear rate were recovered from relations valid for developed steady state laminar flow in a long horizontal pipe [13],

$$\tau = \frac{\Delta P}{2L} r, \tag{1}$$

$$\dot{\gamma} = -\frac{\mathrm{d}v}{\mathrm{d}r}, \tag{2}$$

viscosity, η, being the ratio of shear stress to shear rate, $\eta = \tau/\dot{\gamma}$. A simple measurement of pressure drop was performed to determine the shear stress profile in the measurement volume. In this work, we determined the velocity gradient from flow profiles measured by pulsed Doppler ultrasonic velocimetry. The velocimeter determined the Doppler frequency shift of a backscattered ultrasonic signal as a function of the range of a scattering particle from the transducer. A unique aspect of this work is that we have not assumed that the velocity profile fits that of any rheological model (as done in [14]). The rheograms we obtained were a true inversion of the measured velocity profile.

We used our patented [12] UDV-based inline rheometer unit that combines the necessary Doppler ultrasound and pressure drop instruments into one package. The Doppler instrument used was computer controlled and incorporated time-variable gain (TVG) allowing automated operation in a wide range of materials. The new device design incorporated advances in electronics and signal processing into a single ultrasonic rheometer suitable for deployment in manufacturing environments.

In this work, we also discuss factors that influence the performance of an UDV-based rheometer. The most important factors are related to limits on the spatial resolution of UDV.

2. Experimental

Viscosity measurements were made a non-Newtonian fluid. The fluid used in the experiments was a 0.1-wt.% solution of Carbopol EZ-1 (from B.F. Goodrich) in deionized water neutralized with sodium hydroxide to form a gel. The fluid was seeded with glass beads ("Ballotini Impact Beads, AH-Spec" from PQ Corporation, with diameters between 45 and 90 μm) to a concentration of 0.032-vol%. The beads served to scatter the ultrasound pulses back to the transducer.

The Carbopol mixture was pseudoplastic. Rheograms were determined with a Paar-Physica "Rheolab MC 1" viscometer, for comparison with those derived from velocity and pressure drop. Measurements were made with a DIN spec Z3 cup (with a radius of 13.56 mm) and bob (with a radius of 12.5 mm) at 21–22 °C, for shear rates between 1.29 and 40.8 s^{-1}. Two samples were examined—one taken from the fluid reservoir before the velocity measurements, the other taken after the velocity measurements. Approximate power law correlations of the Paar-Physica results were: "Before" $\tau(Pa) \approx 6.3949\dot{\gamma}^{0.4325}$ and "after" $\tau(Pa) \approx 7.8782\dot{\gamma}^{0.3761}$ for shear rates in s^{-1}. The speed of sound in the Carbopol solution was measured as a function of the concentration of beads and was found to be approximately

$$c\ (\mathrm{ms}^{-1}) = 1495.3 + (0.9898 * \mathrm{vol\%}). \tag{3}$$

The fluid under test was circulated in a loop constructed of 2.093" (0.05316 m) I.D. plastic tubing, as discussed in ref. [7]. The pressure drop per unit length was measured in the straight section using a pair of flush mounted diaphragm pressure transducers (GP:50 model 280-C-PE-2-FS-GJ, 0-2 psig range). The pressure transducers were positioned approximately 3.69 m apart. The large diaphragms of the transducers were placed tangential to the inner diameter of the pipe, to eliminate errors that would result from blocked pressure ports. The fluid under test was circulated with a progressive cavity pump (Moyno 500 series model 33304) with a variable speed drive. A surge bottle was installed downstream of the pump to dampen pressure fluctuations. For the experiments with Carbopol the volumetric flow rate was measured with a gear-type flow meter (FlowData Inc. model EC05) before the fluid entered the loop. The plastic block holding the ultrasonic transducer was bored out to match the pipe ID to within 0.005 in., taking care to keep the pipe and block concentric. The diameters of the pipe and block have to be matched carefully in order to maintain the fully developed flow profile in the block. A single ultrasonic transducer was installed for the Doppler velocity measurements. The transducer (XACTEX model IM-HP-1/4-5 XIM 1055-27) was inserted into a port in the block that was inclined at an angle of 45° with respect to the centerline of the block. The transducer sat in the port with a volume of stagnant fluid in front of it, its face wetted by the process fluid. It was positioned in the port so that one edge of the face was flush with the wall of the block. The port was on the bottom of the block so that air bubbles would float away from the transducer face.

The drive signal sent to the ultrasonic transducer consisted of a 5 MHz carrier modulated by a pulse train. In order to obtain good range resolution it is essential to transmit a short ultrasonic pulse. The range resolution in the beam direction is approximately equal to one-half of the spatial width of the pulse. The pulses used in this work were short (5–20 cycles of the carrier wide), effectively sampling the Doppler waveform and providing a range resolution of approximately 1 mm. The bandwidth of the modulation is typically from 5–25% of the center frequency. The Doppler frequency is in the order of kilohertz, whereas the bandwidth of the transmitted signal is in the order of megahertz. For this reason, the Doppler shift cannot be detected in a single signal pulse transmit/receive sequence. However, the Doppler frequency can be sampled by transmitting and receiving many pulse modulated waveforms at a fixed pulse repetition frequency (PRF).

In this approach, the PRF has prescribed limits. The maximum PRF for an unambiguous signal is set by the pulse transit time. The minimum PRF is that which will resolve the Doppler frequency for the highest particle velocity (two points per cycle to prevent aliasing, or $2f_D$). Combined, these requirements set the greatest upper bound on the flow velocity that can be measured. For the 2-in. diameter pipe used together with transducer geometry and sonic velocity discussed above, this upper bound on velocity measurement was just over 1 m s^{-1}.

The backscattered sound from each pulse modulated transmission was received, quadrature demodulated (outputting I and Q channels) and range-gated, using approximately 100 ranges across the width of the pipe. The results were saved for long sequences of pulses (of the order of 1024). A complex FFT of $I + iQ$ at a given range over the duration of the pulse sequence yielded a power spectrum that was usually peaked at the Doppler frequency. In this work, the half-energy point of the integrated power spectrum was chosen as the Doppler frequency for a specified range. From this frequency the velocity at that range x was calculated using the usual relationship,

$$v(x) = \frac{c}{2\cos\theta}\frac{f_{\text{Doppler}}(x)}{f_{\text{carrier}}}, \tag{4}$$

where θ was the angle (in this case 45°) of the ultrasonic beam off the pipe axis. Performing the FFT and estimating the Doppler frequency from the spectrum for every range across the width of the pipe yielded the velocity profile. The range data provided by the Doppler velocimeter were corrected for the distance along the beam axis from the center of the active piezo element (which sits behind a thin epoxy impedance matching layer and a small pocket of stagnant fluid) to the inner wall of the block.

The pulse repetition frequencies were chosen to be at least twice the expected maximum Doppler frequency to avoid aliasing. In this work, the repetition rate was set at approximately 5 kHz. The maximum velocity that could be measured at this particular PRF was approximately 0.5 m s^{-1} (this is less than the greatest upper bound discussed earlier). The duration of the entire pulse train was approximately 0.2 s. With this duration the best possible velocity resolution was of the order of +/−1 mm s^{-1}.

In this work, 16 velocity profiles were obtained at each volumetric flow rate (listed in the Section 3). The shear rate profile was represented by a cubic spline

$$\dot{\gamma} = \sum_{j=0}^{N} \alpha_j \varphi_j(r/R), \tag{5}$$

where the φ_j are b-spline basis functions, r is the distance from the centerline, and R is the radius of the pipe. Six basis functions were used ($N = 5$), positioned at equally spaced knots from the centerline of the pipe to the wall. The integral of the shear rate was fit to the 16 velocity profiles to determine the spline parameters α_j. The data were weighted by the estimated scan-to-scan standard deviation of the velocity at each range, and the normal equations (of least squares analysis) were modified to include a slight stiffening of second derivative of the spline at the interior knots to improve stability (the modifying terms introducing the prior knowledge that the shape of a velocity profile is not too different from one for a fluid with a constant viscosity). With this inversion procedure differentiation was avoided and semi-empirical models of the viscosity were avoided. The method can, in principle,

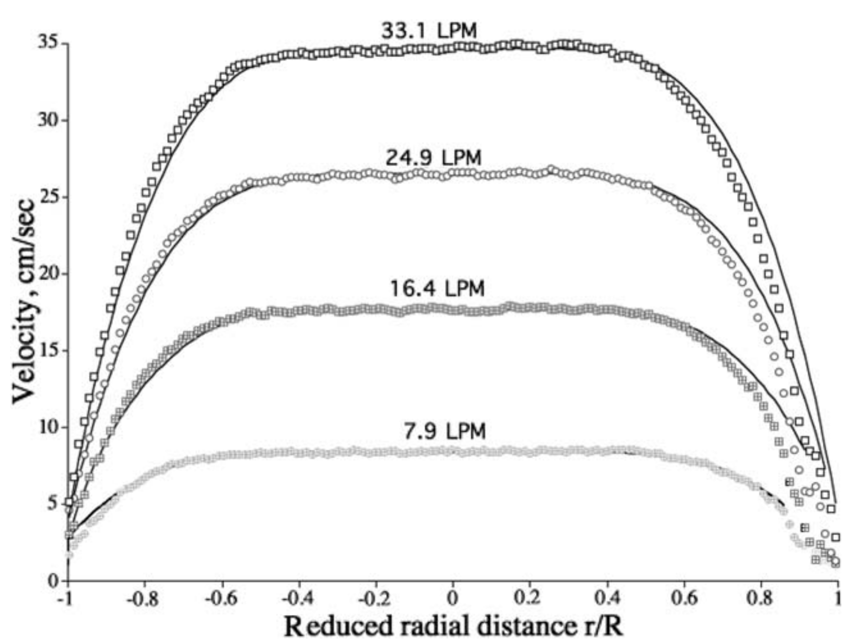

Fig. 1. Measured and fitted velocity profiles in flowing Carbopol solution.

represent Newtonian, Bingham plastic, pseudoplastic or dilatant behavior.

3. Results

Pressure drops and velocity profiles of the Carbopol solution were measured for four cases, at flow rates of 33.1, 24.9, 16.4 and 7.9 $l\,min^{-1}$ (as measured with the gear flow meter), for subsequent processing to obtain the shear rates. Measured and fitted velocities for the four cases, in $cm\,s^{-1}$ are plotted versus reduced radial distance r/R in Fig. 1. The wall nearest the transducer face is located at -1 on the abscissa, the far wall is located at $+1$. The velocity data were weighted during the curve fitting by the scan-to-scan standard deviations. Velocity data were more scattered near the walls, the scatter being greatest near the far wall. It is hypothesized that the increased scatter resulted from a reduction in the peak amplitude of the spectrum near the walls caused by velocity gradient broadening. Near the walls the velocity gradient was large, and the width of the beam samples a broad range of velocities leading to a reduction in peak spectral amplitude. Each

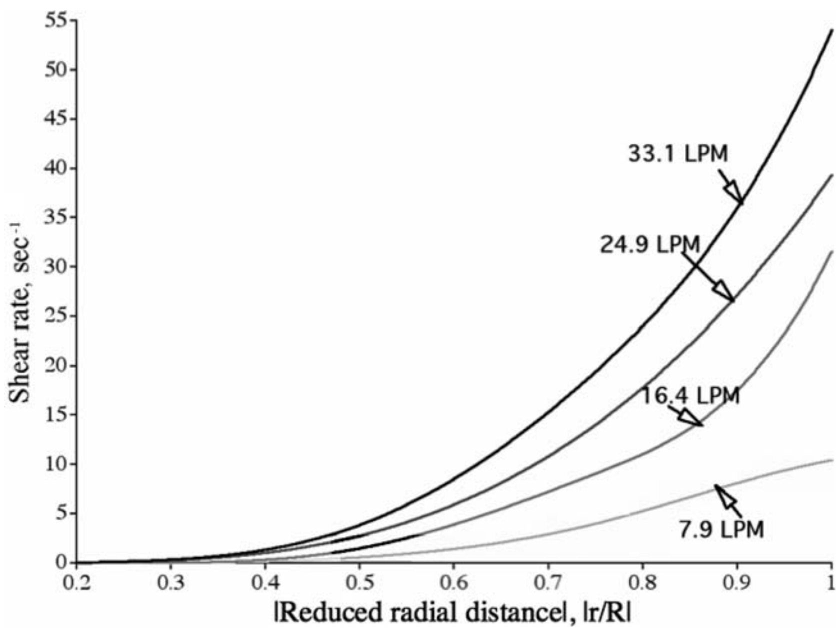

Fig. 2. Calculated shear rates versus position in Carbopol.

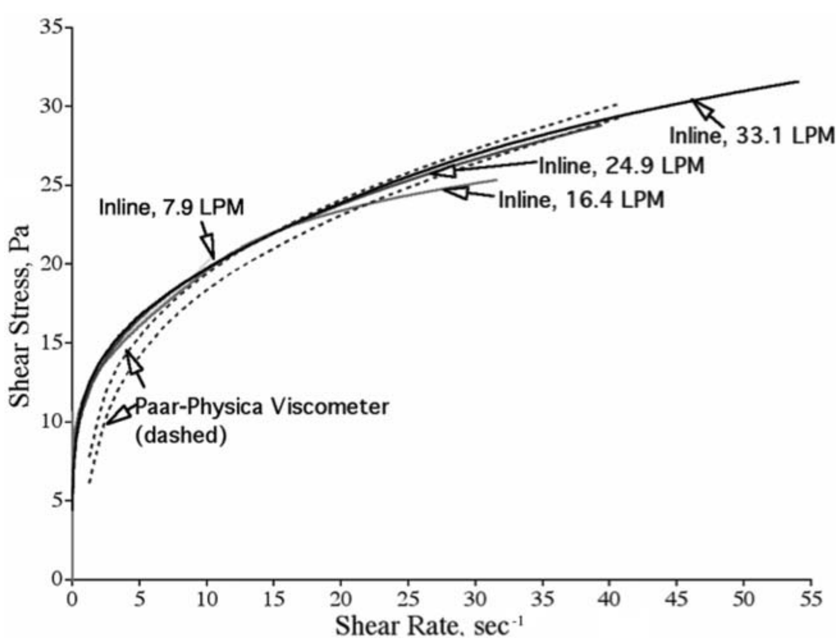

Fig. 3. Shear stress versus shear rate for Carbopol solutions.

fitted velocity profile curve in Fig. 1 lies closer to the near wall data than to the far wall data, because the former were more heavily weighted. Measured velocities near the far wall tended to be lower than those close to the near wall, at the same distance from the centerline. Measured far wall velocities were slightly underestimated, as the velocity gradient broadening shifted the half-energy point of the integrated spectrum to frequencies below those present along the beam axis.

The shear rates derived from the fitted profiles for the four Carbopol cases are plotted versus reduced distance $|r/R|$ from the centerline of the pipe in Fig. 2. They generally behave as expected, except for the two lowest flows with $|r/R| > 0.9$. The two low flow rate cases exhibit some undesirable curvature near the pipe wall. In all cases, scatter in the velocity data in the central plug region made it impossible to distinguish a non-zero slope in the plug.

In general, the calculated rheograms for the Carbopol solution reproduced the results obtained with the Paar-Physica viscometer. Calculated shear stresses are plotted versus shear rate in Fig. 3, together with the results from the Paar-Physica (the latter being shown as dashed lines). The 7.9 LPM case involves only very low shear rates (less than about $10\,\mathrm{s}^{-1}$) and the plotted curve for this case is largely overlapped by the results obtained at the other volumetric flow rates. The agreement of the inversions with the off-line viscometer is good except for shear rates less than about $10\,\mathrm{s}^{-1}$. The low shear rates correspond to flow in the central plug region, where the slope in velocity is small and relatively uncertain. The results were repeatable at the different volumetric flow rates tested.

4. Factors influencing performance

The optimum ultrasonic carrier frequency is governed by a trade-off between obtaining a strong backscattered signal (which favors using higher frequencies) and between reducing attenuation (which favors using lower frequencies). Finding the optimum usually requires bench testing of samples of the fluid of interest or a similar fluid. Our velocimeter allows frequencies to be switched under software control to speed testing.

The minimum measurable velocity is set by the Doppler frequency resolution of the instrument. The best possible Doppler resolution equals the pulse repetition frequency divided by the number of pulses per scan:

$$\Delta f = \dot{n}/N = (\text{data collection time})^{-1}. \tag{6}$$

Longer scans may be taken to improve frequency resolution, limited only by the long term changes in the flow rate, and by data acquisition or computational constraints. However, limits on the spatial resolution of ultrasound measurements cause much greater uncertainties in the determination of the Doppler frequency, as discussed below.

Spatial resolution is limited by several factors:

1. The temporal width W of the pulse. As the pulse is lengthened, the resolution in the beam direction is reduced. However, as the pulse is shortened pulse energy at the carrier frequency is reduced thereby reducing signal strength.
2. The bandwidth of the demodulation filters. If too narrow, the demodulated I and Q signals will spread in time across the range gates. The system must have bandwidth greater that $c/(2\Delta x)$, where Δx is the desired range resolution.
3. The width of the beam. Finite beam width produces both geometric broadening and velocity gradient broadening of the power spectra [15]. The measured power spectrum at each range t_0 is proportional to the normalized distribution of Doppler frequencies $g_{t_0}(\omega)$ in the measurement volume, neglecting attenuation:
$$\|R_{t_0}(\omega)\| \approx \alpha \cdot g_{t_0}(\omega). \tag{7}$$
The measurement volume is roughly a cylinder having the diameter of the beam and a height equaling one half of the spatial width of the pulse ($Wc/2$). The bigger this volume is the broader and lower the distribution is. The distribution of frequencies within the volume is wider near the walls where the velocity gradient is large and becomes wider the larger the ratio of beam to pipe diameter. The frequency at half-energy of a broadened spectrum tends to be lower than the desired Doppler frequency at the midpoint of the measurement volume, introducing a low bias in the velocity measurement.

5. Conclusions

The method presented in this paper provides shear viscosities of a fluid over the whole range of shear rates present in a pipe flow. The method is non-invasive, works on optically opaque fluids, does not interrupt the process and can provide continuous output. It is a true inversion of the pipe flow profile, with only minimal assumptions about the mathematical properties of the shear rate and the kinematics of pipe flow, and no assumed physical model of the rheology. The method provides measurements of viscosity over the whole range of shear rates present in the pipe, under the conditions of shear history actually present in the pipeflow.

Acknowledgements

This work was supported by a grant from the EMSP program of the US Department of Energy. The Pacific Northwest National Laboratory is operated for the US Department of Energy by Battelle Memorial Institute, under contract DE-AC06-76RLO 1830.

References

[1] J. Workman Jr., K.E. Creasy, S. Doherty, L. Bone, M. Koch, A. Ullman, D.J. Veltkamp, Anal. Chem. 73 (2001) 2705.

[2] J. Workman Jr., D.J. Veltkamp, S. Doherty, B.B. Anderson, K.E. Creasy, M. Koch, J.F. Tatera, A.L. Robinson, L. Bond, L.W. Burgess, G.N. Bokerman, A.H. Ullman, G.P. Darsey, F. Mozaynei, J.A. Bamberger, M.S. Greenwood, Anal. Chem. 71 (1999) 121R.
[3] P. Hauptmann, R. Lucklum, A. Puttmer, B. Henning, Sensor. Actuat. A 67 (1998) 32.
[4] R.J. Urick, J. Phys. D: Appl. Phys. 18 (1947) 983.
[5] M. Povey, Semin. Food Analysis 4 (1999) 95.
[6] D.J. McClements, Critical Rev. Food Sci. Nutrition 37 (1997) 1.
[7] A. Shekarriz, D.M. Sheen, in: Proceedings of FEDSM'98 (1998 ASME Fluids Engineering Division Summer Meeting, June 1998), p. 1.
[8] L.C. Lynnworth, G. Jossinet, E. Cherifi, IEEE Ultrason. Symp. (1996) 407.
[9] A. Worch, Meas. Sci. Technol. 9 (1998) 622.
[10] Y. Takeda, Exp. Fluids 26 (1999) 177–178.
[11] D.F. Arola, R.L. Powell, G.A. Barrall, M.J. McCarthy, J. Rheol. 43 (1999) 9.
[12] Patent 6,067,861 for Method and Apparatus for Ultrasonic Doppler Velocimetry Using Speed of Sound and Reflection Mode Pulsed Wideband Doppler.
[13] Y. Uludag, M.J. McCarthy, G.A. Barrall, R.L. Powell, Macromolecules 34 (2001) 5520–5524.
[14] B. Ouriev, E.J. Windhab, Exp. Fluids 32 (2002) 204–211.
[15] P.R. Hoskins, P.J. Fish, S.D. Pye, T. Anderson, Ultrasound Med. Bio. 25 (1999) 391–404.