

INVESTIGATION OF THERMAL CONDUCTIVITY OF A POLYMER SOLUTION AS FUNCTION OF SHEARING RATE

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ABSTRACT

A novel research apparatus is developed to measure the fluid thermal conductivity while in shearing flow, and to determine its dependence on the shearing itself, contrary to the current state-of-the-art of measuring thermal conductivity under the condition of motionless fluid.

A concentric cylinders' apparatus was developed to provide controlled heat transfer in the radial direction, orthogonal to the circumferential fluid velocity, thus virtually preserving pure conductive heat transfer mode. The measurement and control are accomplished and integrated by using a computerized data acquisition system and a comprehensive virtual instrument, developed using the LabVIEW application software.

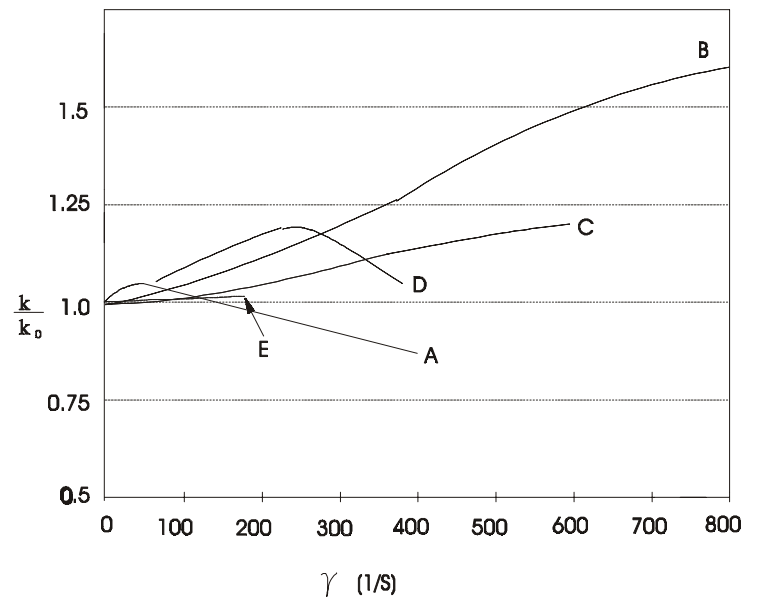
It was found that the thermal conductivity of a Newtonian fluid, such as distilled water, was virtually independent of the fluid motion, as expected. However, for non-Newtonian fluids such as 1000 and 2000 wppm aqueous polyacrylamide (Praestol) solutions, there was up to 10-20% increase of thermal conductivity in the operating shear rate range ($40 \leq \dot{\gamma} \leq 510 \text{ sec}^{-1}$) at 27°C average fluid temperature.

1. INTRODUCTION AND BACKGROUND

It is known that high molecular polymeric solutions and other rheologically complex non-Newtonian fluids are affected by shearing flow: becomes fiber-like, non-uniform and non-isotropic. An innovative method and a novel research apparatus are developed to measure the thermal conductivity of a fluid while it is subjected to shearing flow, thus measuring the thermal conductivity as a function of temperature and shearing parameters themselves (Tong, 1997; Kostic, 1998; Kostic and Tong, 1999). Such measurements are essential because the fluid (changing) structure and anisotropy are flow-induced and dependent. To increase control of the parameters and accuracy, the flow should be isometric (laminar and one-dimensional) and heat

transfer should be only in the transverse direction to the fluid velocity, i.e., orthogonal to it, to prevent interference from convective heat transfer.

Only a few previous measurements, investigating the effect of shear rate on the thermal conductivity of viscoelastic polymer fluids, have been found. Furthermore, the limited



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| A | Polyethylene melts at 150 °C | Picot at al. (1982) |
| B | CMC 1500 wppm at 50°C | Lee (1995) |
| C | Separan 1000 wppm at 30°C | Lee (1995) |
| D | Dow 200 fluids at 25 °C | Chitrangad and Picot (1981) |
| E | 6000-CS at 27 °C | Cocci and Picot (1973) |

Fig. 1: Qualitative curves of the shear-rate-dependent thermal conductivity of non-Newtonian fluids as reported in the literature.

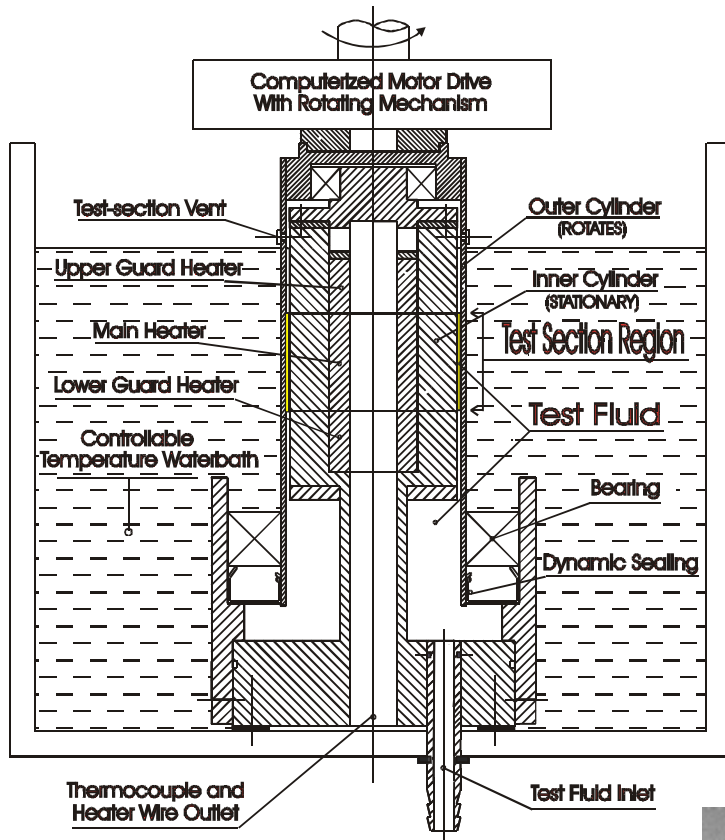


Fig. 2: A novel thermal conductivity apparatus

results have been contradictory as seen on Figure 1, on which the qualitative curves of shear-rate-dependent thermal conductivity of non-Newtonian fluids, previously reported in the literature, are shown. Picot et al. (1982) claimed that the polymeric materials, due to their structure, generally show increasing thermal conductivity with the molecular weight, in both the solid and liquid state. This has been confirmed for solid polymers by Hellwege and Knappe (1963) and for semicrystalline polymers by Gibson et al. (1977), and Choy et al. (1978 and 1980). Polymer solutions in shear flow also exhibit this behavior, that the thermal conductivity of viscoelastic polymer fluids is dependent on shear rate (Cocci and Picot, 1973). Chitrangad and Picot (1981) have interpreted the thermal conductivity anisotropy as being caused by preferred molecular orientation present in the shear flow situation. Picot et al. (1982) reported that, the polyethylene melts at 150°C, exhibited a 2 % increase at a shear rate of 50 1/s, followed by a gradual decrease until a value of 10 % below the zero-shear-rate thermal conductivity at 400 1/s shear rate. In addition, Chitrangad and Picot (1981) showed that the thermal conductivity of Dow 200 at 25°C increased about 13% over a shear rate region ($60 \leq \dot{\gamma} \leq 240$ 1/s) and then decreased over a shear rate region

($240 \leq \dot{\gamma} \leq 380$ 1/s). Furthermore, Wallace et al. (1985) showed that the thermal conductivity of polyethylene melts either increased or decreased with shear rate depending on the molecular weight. Cocci and Picot (1973) found a 10% thermal conductivity increase over a shear rate region ($0 \leq \dot{\gamma} \leq 200$ 1/s) for the 6000-CS (silicone fluid) at 27°C. Finally, Lee (1995) showed that for non-Newtonian fluids such as aqueous CMC and polyacrylamide (Separan) solutions, there was significant increase in thermal conductivity of up to 70% for CMC and 50% for Separan depending on the shear rate, polymer concentration, and temperature. In addition, Loulou et al. (1992) and Chaliche et al. (1994), respectively, reported that the smaller the concentration of the polymer solution, the larger the change of thermal conductivity.

Because the understanding about the shear-rate-dependent thermal conductivity of non-Newtonian fluids is very limited, it will require a lot of research to resolve this problem. The main purpose of this study was to develop a novel thermal conductivity measurement apparatus for investigation of fluid thermal conductivity as function of shearing itself and thus to contribute to better understanding of the phenomena.

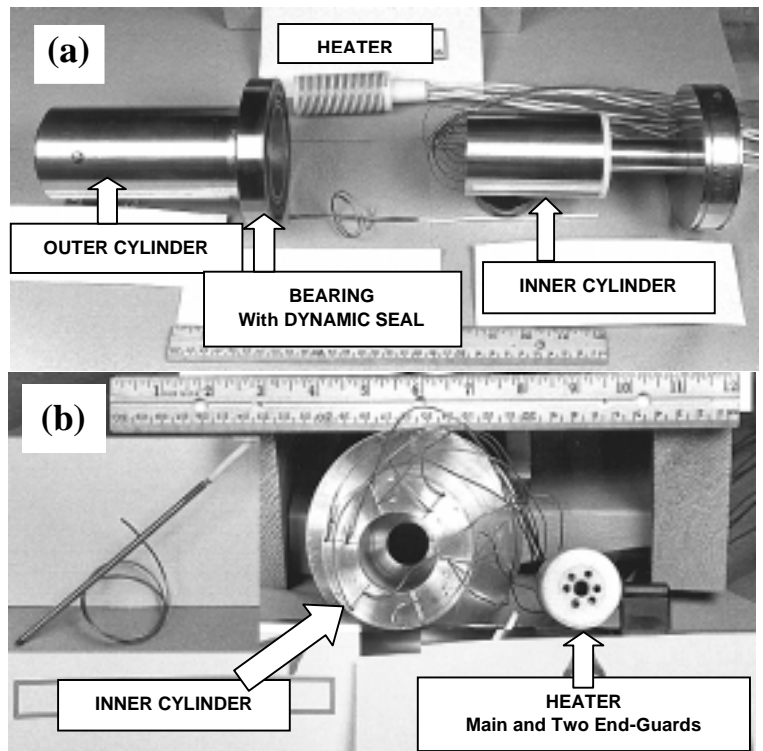


Fig. 3: Photograph of outer cylinder with bearing, heater element, and inner cylinder (a); top views of inner cylinder with thermocouples and heater element (b).

2. INNOVATIVE THERMAL CONDUCTIVITY APPARATUS

A detailed description of the mechanical design is given elsewhere (Tong, 1997; Kostic and Tong, 1999). The apparatus, see Figs. 2 and 3, consists of: (1) an innovative, concentric-cylinders thermal conductivity cell; (2) a high performance, variable controlled-voltage or -current, DC power supply for the main heater; (3) two common, variable-voltage, AC power supplies for the guard heaters; (4) a variable speed DC motor with drive and controller; (5) a constant temperature bath controlled by a high performance, digital immersion circulator; and (6) computerized data acquisition system with signal conditioning hardware and LabVIEW application software (Kostic, 1998). Brief descriptions of selected important components and functions are given below.

Thermal Conductivity Cell:

The actual geometry of an apparatus and test fluid sample consists of a circumferential narrow gap (see Fig. 2), similar to the apparatus for viscosity measurements (Jimenez and Kostic, 1994). In addition, the appropriate heat transfer flux in the transverse to test fluid flow direction is provided. The main test-section dimensions are: $D/d = 2.598/ 2.488$ inch ($=65.99/63.20$ mm), outer/inner cylinder diameters respectively, with the 0.055 inch (1.397 mm) thick gap, filled with the test-fluid in-between. The inner-cylinder's in-the-test-fluid immersion length is 3.8 inch (96.52 mm). It is heated by three 1.3 inch (33.0 mm) diameter electrical-resistance heaters, the central main heater with height $h=1.44$ inch (36.6 mm), and the two remaining guard heaters of 0.78 inch (19.8 mm) high each. The inner cylinder with the heaters assembly is stationary, while the outer cylinder rotates (thus suppressing the Reynolds vortices) generating the Couette type-laminar flow of the test fluid. The two guard-heaters are controlled in such a way to maintain uniform axial temperature in the central, main-heater region, so that the latter heat flux is virtually in the radial direction only. Due to absence of the test-fluid's radial and axial velocities in the main-heater test-section region, the heat flux through the test fluid there is virtually transferred by conduction mode only. Thus, the measurement of the test-fluid's thermal conductivity, while undergoing shearing flow, is achieved.

The directional fluid thermal conductivity (k) is calculated as the corresponding conductive heat transfer rate (Q_k) passing through the fluid test sample per unit of heat transfer area and per unit of appropriate temperature gradient, as a function of temperature level and the shearing rate:

$$k = C(Q_k/\Delta T) \quad (1)$$

where, $\Delta T = T_1 - T_2$ is the measured temperature difference and C represents an instrument constant depending upon the exact geometry of the thermal conductivity cell. If the constant C is determined from the geometrical characteristic of the fluid layer, the method is *absolute*. If the constant C is *calibrated* by using a standard fluid specimen with a known thermal conductivity, the method is *relative*. The thermal conductivity so determined corresponds to an average temperature level of $T = (T_1 - T_2)/2$. For a thermal conductivity cell of Figure 2 with guard heaters and unidirectional radial heat flow, the constant C may be easily calculated, with the nomenclature given above, as:

$$C = \ln(D/d)/(2\Delta h) \quad (2)$$

Although the formula used to determined thermal conductivity from the measured quantities are simple (Eqs. 1 & 2), one should not be misled, due to the fact that the perfect conditions for which the equations are valid never exist, and many corrections must be applied, for example:

$$Q_k = (E \pm \Delta E)(I \pm \Delta I) \pm Q_r \pm Q_c \pm Q_{L.I} \pm Q_{us} \pm Q_{inh} \pm Q_{ch.r} \pm \dots \quad (3)$$

where the electrical heater power (E-I) must be corrected for radiation (r), free convection (c), lead-in losses ($L.I$), losses in heat flux due to unsteady state conditions (us), inhomogeneities (inh), possible chemical reaction ($ch.r$) between the fluid and the wetted surfaces, etc., respectively. Most of these corrections are very difficult, sometime impossible, to take into account. Therefore, in order to increase the accuracy of the measurements, the design of the thermal conductivity cell is such as to minimize those corrections. For example, the guard heaters will prevent end-effects/heat losses; proper material and surface finish will minimize radiation heat transfer; the thin gap will prevent free convection; accurate dimensions will minimize errors of the constant C ; keeping minimal oscillations of the heating and cooling sources will minimize unsteady effects, etc. Also, due to fluid shearing motion (which may be beneficial - ironically and incidentally), the unwanted motion of the fluid particles in the heat transfer direction, i.e., in the orthogonal to the main (only) flow direction will be suppressed by virtue of the shearing itself.

3. INSTRUMENTATION AND MEASUREMENT

The required variables for thermal conductivity measurement are heat flux and temperature gradient through the test fluid, as well as the shearing rate of the test fluid. The apparatus' instrumentation is described next:

- The thermal conductivity apparatus is instrumented and equipped with twelve thermocouples imbedded in the inner cylinder at two different radial and five different

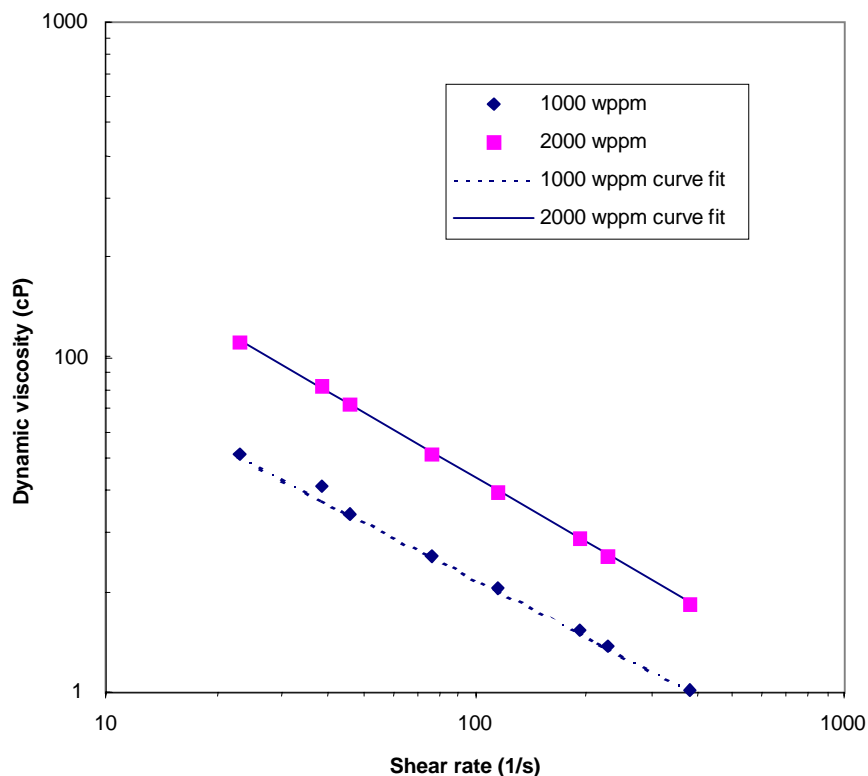


Fig. 4: Dynamic viscosity of Praestol solutions (1000 and 2000 wppm)

axial locations (see Fig. 3; Tong, 1997; Kostic and Tong, 1999). In addition, two thermocouples are attached to the outside surface of the outer cylinder, diametrically opposite at the center of the main-heater’s axial location. These two thermocouples rotate with the outer cylinder and are terminated at its top-end with a quick four-wire connector. Due to this special circumstance, the outer cylinder temperature is measured after the steady state is achieved and after all other measurements are completed, by quick-stopping of the cylinder rotation and with the quick connector wired to the data acquisition system. Then, one more measurement of all other temperatures is performed to confirm the agreement with the corresponding measurements just before the quick stop. Due to the rotation of the outer cylinder, it is not known to the authors if direct measurements of its temperature were done before. Two more thermocouples are used for measurement of constant temperature bath and room temperatures. All thermocouples are made from 30-gauge, T-type thermocouple wire and calibrated before and after assembly. These 16 thermocouples provide for

the temperature gradient calculation needed for thermal conductivity measurement, and for confirming both, the unidirectional (radial only) heat flux through the test fluid and steady state thermal condition.

- The heat flux is measured through measurement of the DC voltage drop across the main heater and a precise current resistor (shunt).
- Finally, the fluid shear rate is calculated using the known test-section geometry and the measured rotational speed of the cylinder with a calibrated tachometer-sensor.

All measurements are repeated until the kinematics and thermal equilibrium is achieved. After that a number of final measurements are performed and results are obtained using statistical analysis, as described elsewhere (Tong, 1997; Kostic and Tong, 1999).

Computerized Data Acquisition and Control

The measurement and control are accomplished and integrated by using a computerized data acquisition system and a comprehensive so called “virtual instrument,” developed using the LabVIEW application software, as described elsewhere (Kostic, 1998). The motor’s rotational speed is measured by a tachometer-sensor and controlled by a voltage-varying DC motor through a built in, solid-state, servo power-amplifier circuitry. The main heater is powered and controlled by a high-quality DC power supply, while two guard heaters are powered by common AC power supplies, and controlled, including over-heating protection, by the computerized system through solid-state relay switches. The computerized system hardware consists of a National Instruments’ MIO plug-in data acquisition board, shielded cable assemblies, and a signal conditioning module with a cold-junction compensated terminal block for thermocouple signals.

Table 1: Rheological properties for power law model				
Solutions (wppm)	Power law model		Power law model	
	<i>n</i>	<i>K</i>	<i>n</i>	<i>K</i>
	(Present values)		[Lee (1995) values]	
Shear rate range	40 < $\dot{\gamma}$ < 400		0.1 < $\dot{\gamma}$ < 200	
1000 wppm Praestol	0.43	0.3012	0.5524	0.2800
2000 wppm Praestol	0.36	0.8354	0.4944	0.5500

NOTE: Lee (1995) was using similar polyacrylamide brand (Separan by Dow Chemical Co.).

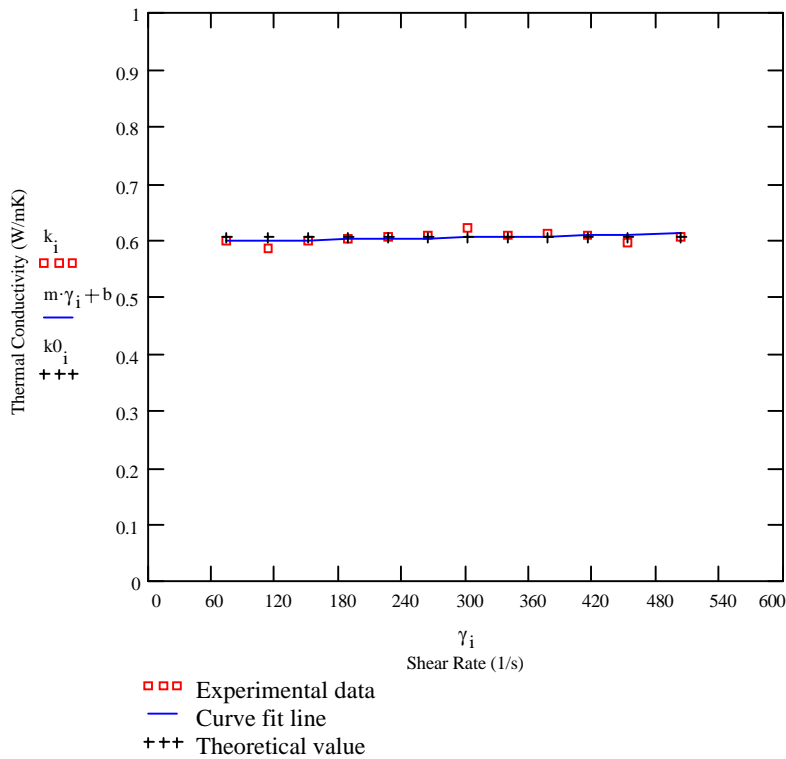


Fig. 5: Thermal conductivity vs. shear rate for distilled water.

4. TEST FLUIDS

Distilled water, as a standard Newtonian fluid with known thermal conductivity, was used for over-all calibration of the apparatus. Then, the thermal conductivity of non-Newtonian fluids, aqueous solutions of polyacrylamide (1000 and 2000 wppm, weight parts per million), was measured as a function of shearing parameters. The polyacrylamide used was manufactured by Stockhausen, Inc., Greensboro, NC, under a brand name Praestol-2273. The aqueous solutions of polyacrylamide were investigated since they were reported to have shear-rate dependent thermal conductivity (Lee, 1995), as well as certain anomalous flow and heat transfer behavior (Kostic, 1994).

Shear Rate Dependant Viscosity Measurements

A digital Brookfield, cone-and-plate viscometer (CPV) was used to measure the dynamic viscosity of non-Newtonian fluids. The CPV, with a calibrated beryllium-copper spring connecting the drive mechanism to a rotating cone, is driven at discrete rotational speeds over its full range. The viscometer was calibrated with a 100 cPoise standard fluid. The measured dynamic viscosity of the Praestol-2273

solutions (1000 and 2000 wppm) are presented on Figure 4, while the calibration data are given elsewhere (Tong, 1997; Kostic and Tong, 1999).

Figure 4 indicates the power law viscosity relation over the region of shear rate measured (about $40 < \dot{\gamma} < 400$ 1/s), for the two Praestol solutions. The power law equation is applicable at this shear rate range, since the log-log curve fit is close to the straight line. The rheological properties, power law index, n , and consistency, K , of the Praestol solutions are presented in Table 1, which includes similar results of Lee (1995) for comparison purposes.

5. THERMAL CONDUCTIVITY RESULTS

Distilled Water Thermal Conductivity

The measured thermal conductivity, as function of shear rate, for distilled water at about 27°C average fluid temperature is shown on Figure 5. The maximum Reynolds number for these measurements were 1300. Note that for the non-Newtonian fluids the maximum Reynolds numbers were lower, about 600 for 1000 wppm, and about 300 for 2000 wppm Praestol aqueous solutions. The water data were obtained using the present conductivity cell and compared with the known

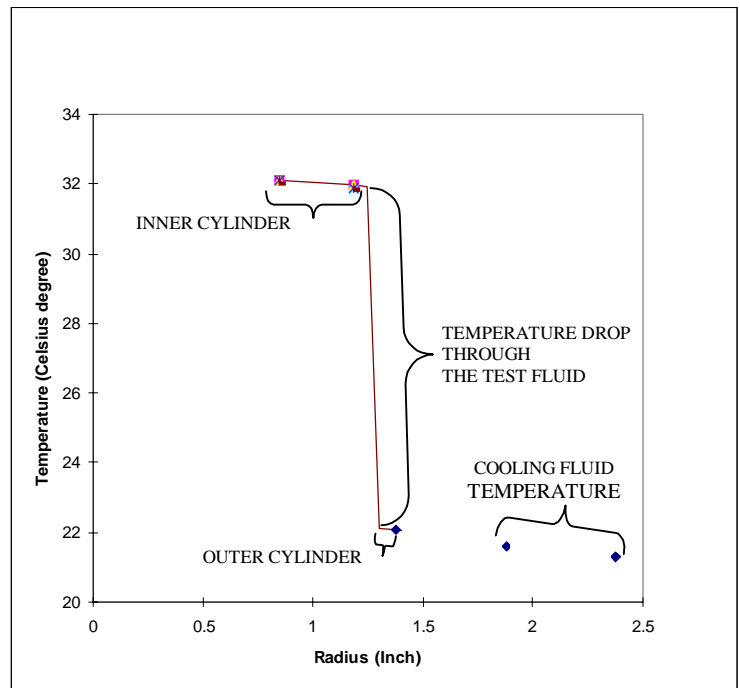


Fig. 6: A typical temperature distribution in radial direction (for 1000 wppm Praestol solution)

values. For the investigated shear rate range, the measured values agreed within 3% difference of the known values of thermal conductivity for distilled water. Also note that there is virtually no shear rate effect on the data, as expected for a Newtonian fluid. These measurements were also used for the purpose of over-all calibration of the apparatus and assessment of its uncertainty (Tong, 1997; Kostic and Tong, 1999).

Thermal Conductivity Measurements for Non-Newtonian Fluids in a Shear Field

Two concentrations of aqueous Polyacrylamide (Praestol 2273) solutions were chosen for investigation. The 1000 and 2000 wppm Praestol-2273 solutions were examined at varying shear rates, from 50 to 510 1/s. A characteristic temperature distributions is presented in Figure 6. The final results of these experiments are presented in Figures 7 and 8, as fluid thermal conductivity plotted against shear rate for an average fluid temperature about 27 °C. The curves in Figures 7 and 8 show an obvious dependence of the thermal conductivity on the shear rate. The thermal conductivity increases more (17%) with shear rate for the lower concentrated 1000 wppm Praestol solution than the 13% increase of the higher concentrated 2000 wppm Praestol solution, which is in qualitative agreement with Lee's (1995) findings. According to Lee (1995), the effect of concentration of polyacrylamide Separan solutions to the variation on the thermal conductivity at a low shear rates ($\gamma \leq 200$) and low temperatures is very weak, but the effect increases with an increase of shear rate ($\gamma \geq 200$).

6. CONCLUSIONS AND RECOMMENDATIONS

A novel research apparatus is developed to measure the fluid thermal conductivity while in shearing flow, and to determine its dependence on the shearing itself, contrary to the current state-of-the-art of measuring thermal conductivity under the condition of motionless fluid.

One of the objectives of this project was also to utilize the latest powerful, yet inexpensive, technological developments: sensors and transducers, computerized data acquisition, and application software (Kostic, 1998). The designed, computerized measurement and data acquisition system, accomplishes the following functions: (1) acquire measured data with high speed and accuracy; (2) interactively process and analyze measured data for immediate use or future post-processing; (3) provide interactive and accurate feed-back process control - motor speed and guard-heating power, and (4) interactively displays the raw/measured and processed/analyzed data in graphical and/or numerical forms. In addition, such a system allows for easy modification and enhancement of so called "virtual (software) instruments" by modification of software programs.

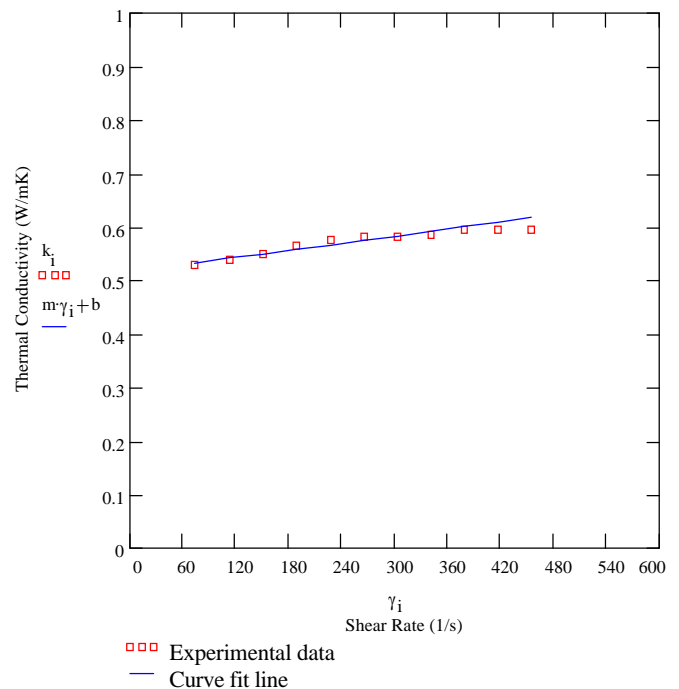


Fig. 7: Thermal conductivity vs. shear rate for 1000 wppm Praestol solution.

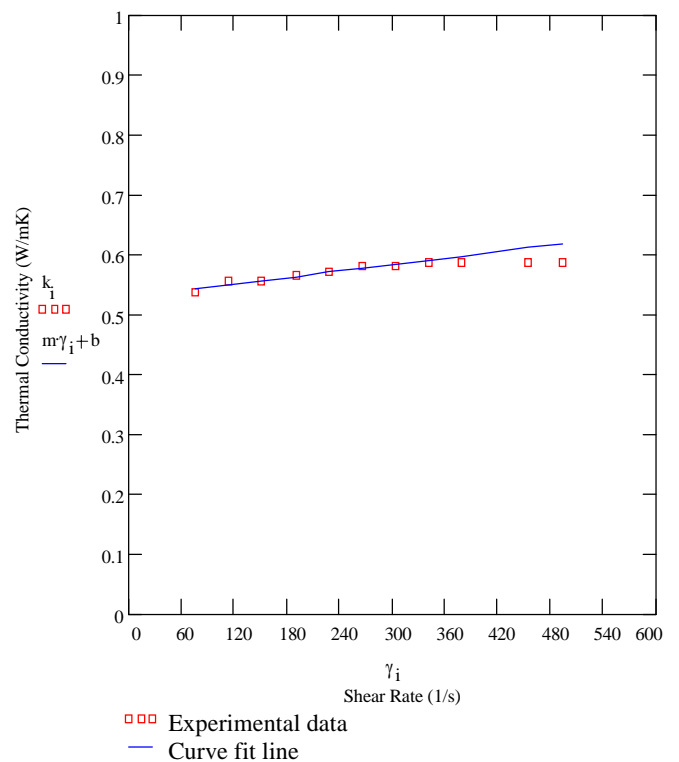


Fig. 8: Thermal conductivity vs. shear rate for 2000 wppm Praestol solution.

It was found that the thermal conductivity of a Newtonian fluid, such as distilled water, was as expected, virtually independent of the fluid motion, and within 3% agreement of the known values. These measurements of known thermal conductivity fluid were utilized for over-all apparatus and procedure calibration and uncertainty assessment, the latter being about 5% for relative (differences) and about 10% for absolute values of the property measurements. However, for non-Newtonian fluids such as 1000 and 2000 wppm aqueous polyacrylamide (Praestol-2273) solutions, there was up to 10-20% increase of thermal conductivity in the operating shear rate range ($40 \leq \dot{\gamma} \leq 510 \text{ sec}^{-1}$) at about 27°C average fluid temperature. These results are in qualitative agreement only with available similar results by Lee (1995), but the present thermal conductivities are quantitatively much smaller than the limited Lee's data. Due to complexities of the phenomena and difficulties involved in investigation, further improvements of the apparatus, instrumentation and procedure, as well as much more measurements for different fluids and temperature levels, and over wider range of shear rates, are needed.

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"Reliable data are the pure aspect of research.. The flaws are in the interpretation."