IN SITU RHEOLOGICAL MONITORING OF VISCOUS NON-NEWTONIAN FLUIDS USING A HELICAL RIBBON AGITATOR

P. J. Cullen[a], A.P. Duffy[a], C.P. O'Donnell[a,]*, D.J. O Callaghan[b].
[a] Department of Agricultural & Food Engineering, University College Dublin, Earlsfort Terrace, Dublin 2, Ireland.
[b] Food Ingredients Department, Dairy Products Research Centre, Teagasc, Moorepark, Fermoy, Co.Cork, Ireland.
*Corresponding author. Fax: + 353 1 475 2119. E-mail: colm.odonnell@ucd.ie

Abstract
Mechanically degraded food fluids were modelled as Power law fluids in a pilot scale (42 l) helical ribbon agitation vessel. Effective rheological representative flow curves determined from speed/torque data proved comparable to off-line laboratory rheometer measured data. Modelling effective viscosity as a Power law function gave effective consistency coefficient K_{eff} which correlated strongly with K from the rheometer (R^2=0.99). Effective flow behaviour index n_{eff} was comparable in magnitude (average 5% difference) to rheometer measured equivalents. Determination of absolute consistency coefficient K using the mixer viscometer technique proved problematic with large differences in magnitude. Single point measurements of raw mixer torque data at a constant speed correlated well against reference off-line techniques, namely the Bostwick Consistometer and consistency index from the rheometer (R^2=0.98 and R^2=0.99), coupled with low standard errors of estimate (4% & 3.2%) respectively. The helical ribbon system evaluated was capable of monitoring the rheological properties of fluids with large particulates.

Keywords: mixing, rheology, process control, non-Newtonian fluids, helical ribbon, food processing

1. Introduction

Increased monitoring of foods at processing stage would allow real-time determination of quality attributes facilitating improved process control. In many process applications final product quality and process efficiency is a function of rheology of the fluid, which will influence such characteristics as pumpability, pourability and spreadability (Cullen, Duffy & O'Donnell, 2001). Quality control rheological measurements have traditionally been carried out on tomato-based products intermittently and off-line using such instruments as the Bostwick Consistometer (Cullen, Duffy & O'Donnell, 2001) and simple dip in viscometers such as the Brookfield viscometer (Barnes, 2001). The Bostwick Consistometer (BC) is a simple, low cost instrument used to monitor product consistency in a range of foodstuffs. However limitations include: operator variability and subjectivity, levelling and dryness of the instrument and serum separation at the edge of product flow, and it is not suitable for on-line consistency measurements. Laboratory viscometers are also widely used as quality control instruments based on simple rotational viscometry, with shear rate dependent upon spindle type and rotation speed used. Replacing this instrument and the Bostwick Consistometer or relating their measurements to a process viscometer would provide continuous real time monitoring during processing.

Numerous viscometers are available for process control, including tube, rotational and vibrational viscometers, suitability of such techniques have recently been reviewed for the food industry (Cullen et al. 2000). Such conventional viscometer designs may prove problematic when dealing with complex food fluids (multiphase, fibrous, particulate and highly viscous), resulting in errors due to phase separation, fouling of measuring gap or wall slip. The ideal viscometer design should facilitate, ease of cleaning in place with minimal possibility of fouling, a quick response time and good sample renewal to ensure the any measurement obtained is representative.

Numerous unit operations in industry require effective mechanical agitation or mixing of fluids. The geometry of agitator used will depend upon properties, particularly viscosity, of the fluid to be mixed. Helical ribbon agitators are frequently used for highly viscous non-Newtonian fluids, providing efficient mixing at the vessel wall and vertical mixing of the fluid (Rai, Devotta & Rao, 2000), with operating speeds generally within the range 30 - 100 rpm. In-situ rheological evaluation of batches, post mixing and prior to pumping, would facilitate improved process control. Benefits include no necessity for bypass processing lines or additional instrumentation in contact with the product coupled with real time monitoring. Use of mixer viscometry, where high precision measurements of torque and impeller rotational speed for a given geometry can determine apparent viscosity in the mixing vessel, is well documented (Cantu-Lozano, Rao & Gasparetto 2000; Ford & Steffe 1986). However these trials are based upon scale down mixing geometries, relying on traditional off-line rheometer/viscometer measurement of torque and speed (Cullen, et al. 2000). Due to the complex flow patterns, and hence shear rates, in mixing vessels only approximate data and average values can be determined (Castell-Perez & Steffe, 1992). The development of a novel low cost non contact torque transducer (Torqusense Sensor Technology Ltd, Banbury, Oxon, UK) providing precise dynamic measurements of rotary torque over ranges covering 0-10 mN.m to 0-10,000 N.m. will assist technology transfer of mixer viscometry to production scale. Principle of operation of this torque transducer involves a surface acoustic wave device used as a frequency dependent strain gauge, which measures the change in resonant frequency caused by the applied strain in the shaft. Accurate measurement of shaft rotational speed using a light beam is also recorded.

The objective of this work was to evaluate the potential of a pilot plant scale helical ribbon mixer as a rheological process control technique, using the following criteria:

1. Determine Power law indices using the mixer viscometer technique.
2. Determine effective Power law indices from representative mixer flow curves.
3. Develop correlations between single point mixer torque measurements and off-line reference techniques.

2. Theoretical Considerations

2.1 Mixer viscometer constant

Power consumption for Newtonian fluids within a mixing system may be expressed in terms of power number $P_o = (P/\rho d^5 N^3)$ and mixing Reynolds number $R_e = (\rho d^2 N/\eta)$, which may be empirically related to each other in the laminar flow region ($R_e<10$), by equation (1). Where, A is a geometric constant.

$$P_o = \frac{A}{R_e} \tag{1}$$

For non-Newtonian Power law fluids, viscosity will increase from a minimum value close to the impeller to a maximum value far away from the impeller. Metzner and Otto (1957) suggested that equation (2) may be used for non-Newtonian fluids if an apparent viscosity is evaluated at an average shear rate.

$$\dot{\gamma}_a = k_s N \tag{2}$$

The shear rate constant of proportionality k_s is dependent upon impeller geometry but is usually found to be independent of fluid properties and impeller speed (Shamlou & Edwards, 1985). There are a number of techniques for determining the constant k_s. Comparisons between the different calculation methods are discussed by (Castell-Perez & Steffe, 1992). Rieger and Novak (1973) demonstrated that if equation (2) is valid, a plot of (1-n) vs. $\log_{10}(P/K\Omega^{n+1}d^3)$, results in a straight line of slope – k_s, for the impeller geometry.

2.2 Power Law indices

Shear stress and shear rate are directly proportional to torque and rotational speed respectively. Therefore the flow behaviour index may be determined from a plot of log torque (M) vs log of rotational speed (N) or angular velocity (Ω).

The consistency index K may be determined by applying equation (3) to two test fluids, one with unknown flow properties (x), and one with known properties (y).

$$\frac{M_x}{M_y} = \frac{K_x \Omega_x^{n_x} (k_s)^{n_x}}{K_y \Omega_y^{n_y} (k_s)^{n_y}} \tag{3}$$

2.3 Representative Flow Curves

Representative flow curves for Power law fluids may be developed to facilitate quality control. Absolute rheological data may not be required for quality control, in this case simple representative flow curves may be utilised and compared to reference flow curves, (Steffe, 1996). Effective viscosity is defined in terms of mixing parameters:

$$\eta_{eff} = \frac{M}{\Omega d^3} \tag{4}$$

Effective viscosity is assumed to be directly proportional to apparent viscosity and shear rate directly proportional to angular velocity. Regression analysis of effective viscosity vs. angular velocity results in an effective consistency coefficient, K_{eff} and an effective flow behavioural index, n_{eff}. The value of K_{eff} is unique to both the fluid tested and equipment geometry used. However the values of n_{eff} are numerically the same as those determined in 2.2.

2.4 Single point measurements

Single point measurements of torque at a constant speed may be used as a quality control technique by developing correlations with reference off-line techniques. However care should be taken, as with any single point measurement technique, to ensure that data is representative of the products flow curve (Barnes, 2001).

3. Materials and methods

A pilot scale helical ribbon agitator (42 l) was constructed consisting of a cylindrical dished-bottom stainless steel vessel incorporating a close clearance helical ribbon impeller, whose geometrical configurations are defined in Fig. 1 and listed in Table 1. Torque and speed were measured with a non-contact rotary torque transducer (Torqsense, Sensor Technology Ltd, Banbury, Oxon, UK) in the range 0-6 N.m and logged with the transducer interface module and system software. A variable speed Heidolph (model RZR 2041) stirrer motor (AGB Scientific Ltd, Dublin) was used to vary agitator speed between 35 and 70 rpm. Temperature was measured in the agitator and simulated on the rheometer for each sample.

Newtonian and non-Newtonian fluids (Table 2) were used to determine the mixer viscometer constant for the agitator using the slope method. Well characterised model fluids (Sigma-Aldrich Ireland Ltd, Dublin) were chosen spanning the spectrum of flow behavioural indices. Xanthan gum aqueous solutions (1%, 1.5%, 2%) were used as model fluids for investigating the effects of elasticity on torque.

Commercial ketchup was thickened by adding tomato paste and samples were varied in consistency from Bostwick values of ca. 2.5 to 10.5, by dilution. A pizza sauce containing particulates of up to 5.0 mm in length , (Green Isle Foods, Naas, Ireland) was selected as a fluid displaying more complex characteristics and varied in consistency over a small range of Bostwick values from ca. 4.0 to 6.0, by dilution. Dilution levels were achieved by adding water quantities of ca 1.5% of total sample volume. Prior to measurement these samples were mixed for 30 minutes at 20 °C. The resulting mechanically degraded samples, simulating post-mixing conditions, were assumed time-independent and modelled as Power law fluids. Off-line consistency measurements were taken for each dilution with a Bostwick Consistometer (Christison Scientific, Gateshead, Tyne & Ware, UK), values were taken as the distance in centimetres the sauce travelled in 30 seconds. Shear rate sweeps (0-300 s^{-1}) were preformed on a laboratory rheometer (Carrimed $CSL^2$100, TA Instruments, Leatherhead, Surrey, UK) using a cone and plate geometry (4 cm & 2° acrylic cone) for tomato ketchup samples and a parallel plate geometry (4 cm & 1500 μm gap) for pizza sauce samples, at batch temperature. BC and rheometer measurements (shear rate sweeps) were performed in triplicate and averaged for each dilution.

4. Results and discussion

4.1 Mixer viscometer constant

Linear regression analysis of the semi-logarithmic plot of (1-n) vs. ($P/K\Omega^{n+1}d^3$), (Fig. 2) yielded a good measure of fit (R^2=0.99). The shear rate proportionality constant k_s was determined from the slope of the line as 10. This constant was determined to be independent of angular velocity for speeds of 40, 50, 60 and 70 rpm with similarly high measures of fit.

4.2 Power Law indices

The flow behavioural index determined from the logarithmic plots of mixer torque vs rotational speed, resulted in comparable results to those measured with the laboratory rheometer, with an average percentage difference in magnitude of 5%. They also characterised the transition of the fluids tested towards more Newtonian behaviour over a small range of dilutions (Fig. 3). These results show that flow behavioural indices may be estimated in-situ using agitators, by measuring torque as a function of rotational speed.

Fig. 4 compares K values, determined using equation (3) to their rheometer measured equivalents for a range of dilutions. Considerably higher consistency index values were obtained with the mixer viscometer, with the discrepancy decreasing with increased dilution. It is suggested this discrepancy is due to the solid fraction of tomato products, resulting in network structure or gel-like behaviour (Rao &

Cooley, 1992). Fig. 5 investigates the viscoelastic behaviour of a ketchup sample, G*′* was higher than G*′′* at all frequencies (ω) measured, indicating that the sample's properties were more solid like than viscous. The decrease in complex viscosity η^* shows the shear thinning nature of the sample. From a structural perspective, log plots of G*′* or G*′′* vs log ω of weak gels yield positive slopes and true gels zero slopes. Thus, it may be concluded that the tomato ketchup tested displayed weak gel like behaviour.

The effect of elasticity on mixing with agitators including helical ribbons is ambiguous, with various authors reporting decreases, increases and no effects on torque (Castell-Perez and Steffe, 1992). However elasticity will produce differences in the flow fields around the mixing impeller and generally it is concluded that elastic properties of fluids tend to reverse secondary flows induced by centrifugal force, (Castell-Perez and Steffe, 1992). A number of authors have reported a clear increase in torque due to elasticity for helical ribbons (Brito *et.al.* 1991; Collias and Prud'homme, 1985). The latter authors reported a tripling of torque for some fluids, due to elasticity. This increase in torque is not a function of the Power law consistency index or flow behavioural index.

To evaluate this theory further, a well-characterised reference fluid, xanthan gum was circulated in the helical ribbon agitator. This fluid exhibits a gel like structure at concentrations greater than 1.0%. The percentage difference in magnitude between the mixer viscometer determined consistency index and its rheometer measured equivalents increased from 3% at the 1% concentration level to 7% and 50% at the 1.5% and 2% concentrations respectively. Therefore, the classical approach for non-Newtonian fluids developed by Metzner and Otto (1957) to determine power consumption in agitator systems, fails to account for the effects of elasticity on torque developed for complex fluids such as tomato sauces. Similarly equation (3) does not account for this effect. Evaluation studies on the effects of elasticity on mixing have used elastic fluids with a constant viscosity (boger fluids). Since most viscoelastic fluids also exhibit strong shear thinning characteristics, power consumption changes may be due to changes in either viscosity or elasticity, (Castell-Perez & Steffe, 1992). The development of a correlation, which includes elasticity and accounts for changes in viscosity and impeller geometry will prove problematic. It is also evident from this work that the Metzner and Otto correlation may not be valid for scaling-up on the basis of power consumption per unit volume, as reported be Oliver et al. (1984), due to the different power requirements as a result of varying fluid elasticity. The effects of elasticity on measured torque are dependent on scale, which may explain why such effects are not observed in the laboratory scale studies reported in the literature (Ford & Steffe, 1986; Castell-Perez & Steffe, 1992).

4.3 Representative Flow Curves

Effective viscosity as determined using equation (4) and plotted against angular velocity was modelled as a Power law function. These representative flow curves are shown in Fig. 6 for a series of dilutions for tomato ketchup samples. Regression analysis of the data resulted in an effective consistency coefficient, K_{eff} and an effective flow behavioural index n_{eff}. Effective consistency correlated strongly ($R^2 = 0.99$) against K measured by the rheometer (Fig. 7), showing the technique may be used for process measurements of fluid consistency. Effective flow behavioural indices (n_{eff}), were numerically the same as those measured above. An advantage of this technique is that no calibration model fluids are required. Also a flow curve is produced for the product, rather than using many quality control procedures which may only provide single point measurements, resulting in a more detailed rheological characterisation. Effective consistency values correlated strongly ($R^2 = 0.97$) against Bostwick Consistometer values.

2.4 Single point measurements

Raw torque data from the agitator correlated strongly ($R^2 = 0.99$ & $R^2 = 0.98$) with consistency index from the rheometer and Bostwick values respectively using a Power law model (Fig. 8). These high measures of fit coupled with low standard errors of prediction (expressed as a percentage of the range), less than 3 and 4 % respectively, show that torque readings at a constant speed may be used to accurately predict either off-line reference measurement. A similarly high measure of fit ($R^2 = 0.98$) was obtained for the correlation developed between the torque readings and Bostwick values for the pizza sauce samples, over a very small consistency range, BC values 4.0 – 6.0 cm. BC measurements were in increments of 0.5 cm, the limit of the instruments sensitivity as reported by Cullen, Duffy & O’Donnell, (2001). Development of a correlation with consistency coefficient, K, as determined from rheometer shear rate sweeps proved more problematic, due to the rheometer's inability to cope with samples containing large particulates, even using the parallel plate geometry with a large gap.

The torque transducer proved sensitive to small changes in product consistency, which is representative of fluctuations in industry and capable of coping with large particulates. If a number of products or recipes are to be evaluated, such correlations should be developed for each specific product. Quality control personnel are generally only interested in knowing if the product produced is within a specified range for day to day production, this range developed previously upon knowledge of the complete flow curve (Barnes, 2001). This quality control range, whether determined by consistometers or single viscosity readings from laboratory viscometers, may be predicted accurately or replaced by mixer torque measurements, which also provides the benefits of real-time process monitoring.

5 Conclusions

Suggested mixer viscometry techniques, coupled with a novel torque transducer were evaluated as a process control tool to monitor rheological properties of fluid foods in an helical ribbon agitator. Post mixing rheological evaluation of tomato based products using representative flow curves within a pilot scale helical ribbon agitator, proved an effective technique to obtain process viscometry measurements. Modelling effective viscosity as Power law functions gave comparable data to rheometer measured results. Reference off-line measurement techniques can be predicted by developing correlations with constant speed torque measurements from the agitator. The torque transducer employed proved sensitive to small changes in product consistency. Absolute Power law indices could not be determined by comparing torque readings using equation (3) to reference non-Newtonian fluid data, due to elasticity variations. The effects of which are dependent not only upon the fluid's viscoelastic properties but also the geometry and scale of operation.

Design advantages of the proposed viscometry system for industry include its ability to cope with complex fluid characteristics such as particulates and fibres. Subjectivity and errors due to operator variability, associated with certain off-line instruments, are also removed. Process monitoring is in real time, which facilitates improved process control. Rheological measurement problems due to slip or particle blockages are not encountered. No additional cleaning or instrumentation in contact with the food product is required. It is suggested that other low speed impeller designs, which operate in the laminar flow region such as anchors and paddles, could also be used for other viscous fluids.

Nomenclature

A	geometric constant
c	clearance between impeller tip and vessel wall (m)
d	impeller diameter (m)
G'	storage modulus (Pa)
G''	loss modulus (Pa)
K	consistency coefficient ($Pa.s^n$)
K_{eff}	effective consistency coefficient ($Pa.s^n$)
k_s	shear rate constant of proportionality
M	torque (N.m)
n	flow behaviour index (dimensionless)
n_{eff}	effective flow behavioural index (dimensionless)
N	impeller rotational speed (rev/s)
P	power input (N.m/s)

Greek symbols

$\dot{\gamma}_a$	average shear rate (s^{-1})
ρ	fluid density (kg/m^3)
η	Newtonian viscosity (Pa.s)

η_{eff}	effective viscosity (Pa.s)
η^*	complex viscosity (Pa.s)
Ω	angular velocity of rotation (rad/s)
ω	angular frequency (rad/s)

Dimensionless numbers

P_o	power number, $P/\rho N^3 d^5$
R_e	mixing Reynolds number, $\rho N d^2/\eta$

Sub-scripts

x	Fluid with unknown flow properties
y	Fluid with known flow properties

References

Barnes, H.A. (2001). An examination of the use of rotational viscometers for the quality control of non-Newtonian liquid products in factories. *Applied Rheology*, 11, 89 – 101.

Brito, E., Leuliet, J.C., Choplin, L. & Tanguy, P.A. (1991). On the role of elasticity on mixing with a helical ribbon impeller. *Trans. I. Chem.E.* 69, 324-331.

Cantu-Lozano, D., Rao, M.A. & Gasparetto, C.A. (2000) Rheological properties of noncohesive apple dispersion with helical and vane impellers: effect of concentration and particle size. *J. Food Process Eng*. 23, 373-385.

Castell-Perez, M.E. & Steffe, J.F. (1992). Using mixing to evaluate rheological properties. In M.A. Rao, & J.F. Steffe, *Viscoelastic properties of foods* (p. 247-284). London: Elsevier Applied Science.

Collias, D.J. & Prud'homme, R.K. (1985). The effect of fluid elasticity on power consumption and mixing times in stirred tanks. *Chem. Eng. Sci*. 40(8), 1495-1505.

Cullen, P.J., Duffy, A.P., O'Donnell, C.P. & O'Callaghan, D.J. (2000). Process viscometry for the food industry. *Trends Food Sci. Technol.*, 11(12), 451-457.

Cullen, P.J., Duffy, A.P., O'Donnell, C.P. (2001). On-line rheological characterization of pizza sauce using tube viscometry. *J. Food Process Eng*. 24 (3), 145-159.

Ford, E.W. & Steffe, J.F. (1986). Quantifying thixotropy in starch-thickened, strained apricots using mixer viscometry techniques. *J. Texture Studies*, 17, 71-85.

Metzner, A.B. & Otto, R.E. (1957). Agitation of non-Newtonian fluids, A.I.Ch.E.J. 3(1) 3-10.

Oliver, D.R. Nienow, A.W. Mitson, R.J. & Terry, K. (1984). Power consumption in the mixing of boger fluids. *Chem. Engng. Res. Des*., 62, 123-127.

Rai, C.L., Devotta, I. & Rao, P.G. (2000). Heat transfer to viscous Newtonian and non-Newtonian fluids using a helical ribbon agitator. *Chem. Eng. J*. 79, 73–77.

Rao, M.A. & Cooley, H.J. (1992). Rheological behaviour of tomato pastes in steady and dynamic shear. *J. Texture Studies*, 23, 415-425.

Rieger, F & Novak,V. (1973). Power consumption of agitators in highly viscous non-Newtonian liquids. *Trans. Instn. Chem. Engrs*. 51, 105–111.

Shamlou, P.A. & Edwards, M.F. (1985). Power consumption of helical ribbon mixers in viscous Newtonian and non-Newtonian fluids. *Chem. Eng. Sci*. 40(9), 1773-1781.

Steffe, J.F. (1996) *Rheological methods in food process engineering* (2nd ed.). Michigan: Freeman Press.

Fig. 1. Schematic of pilot plant helical ribbon agitator.

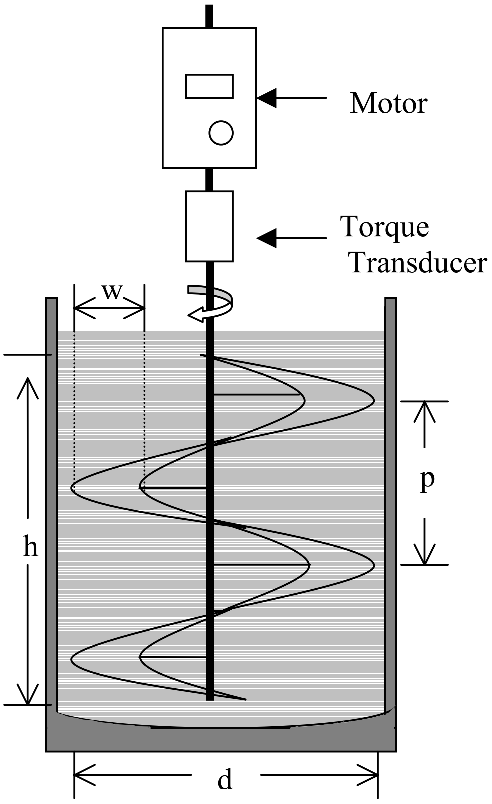

Fig. 2. Semi-logarithmic plot of dimensionless functions $(P/K\Omega^{n+1}d^3)$ vs (1-n) for standard fluids as listed in Table 2.

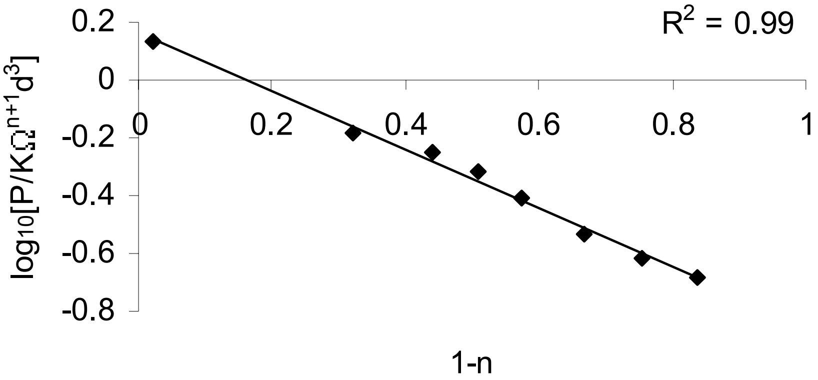

Fig. 3. Logarithmic plot of torque vs rotational speed, for tomato ketchup samples.

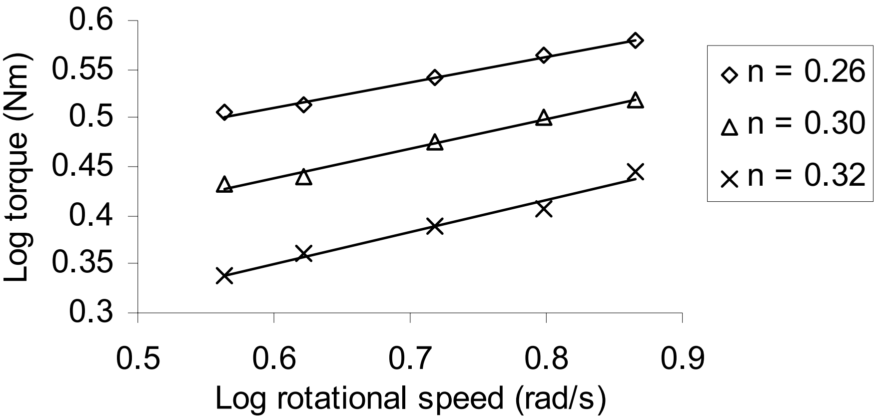

Fig. 4. Consistency index (K) as determined by the mixer viscometer, compared to their rheometer measured equivalents, for tomato ketchup samples (BC values 4.2 - 6.8).

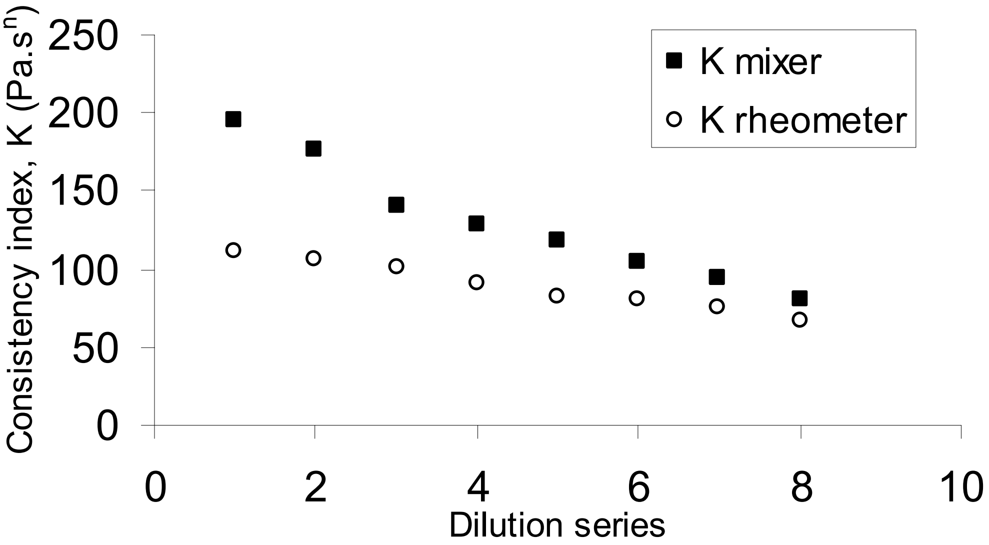

Fig. 5. Logarithmic plot of angular frequency (ω) vs storage (G′) and loss (G′′) moduli and η (η*) for a tomato ketchup sample.

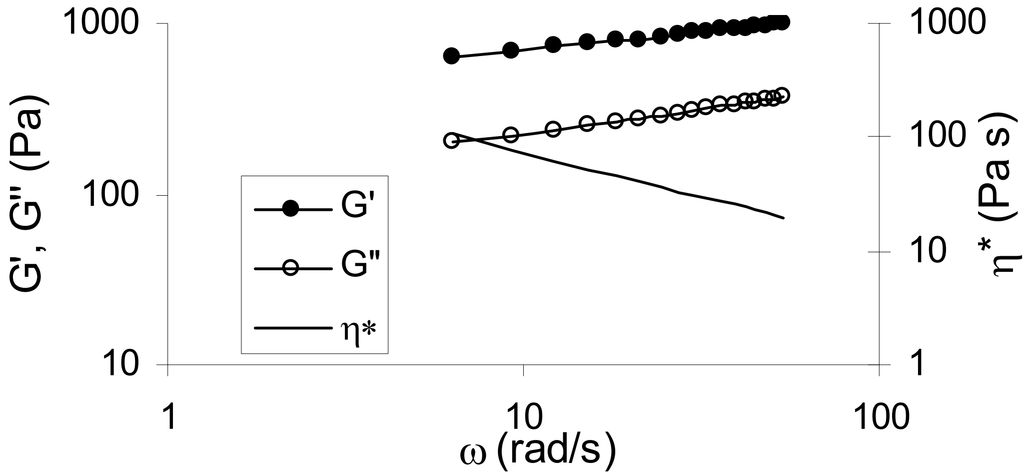

Fig. 6. Representative flow curves of tomato ketchup samples for a series of dilutions (BC values 4.2 – 6.5).

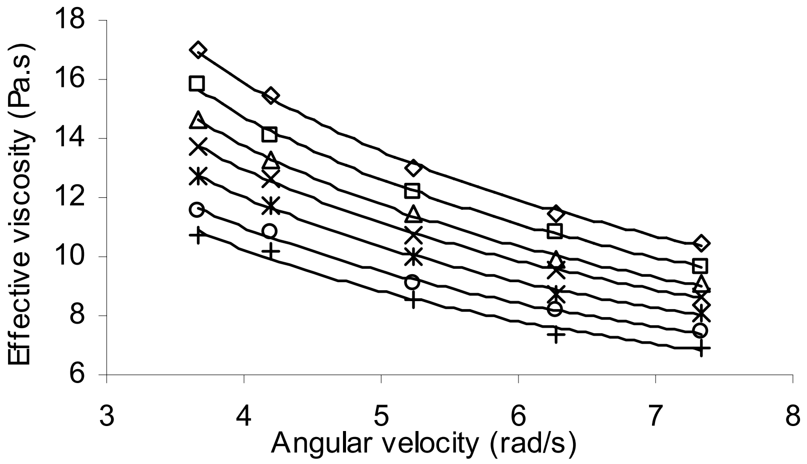

Fig. 7. Linear correlation between effective consistency index (K_{eff}), as determined from the mixer, against rheometer measured consistency index values (K) for a series of dilutions of tomato ketchup samples (BC values 4.2 – 6.5).

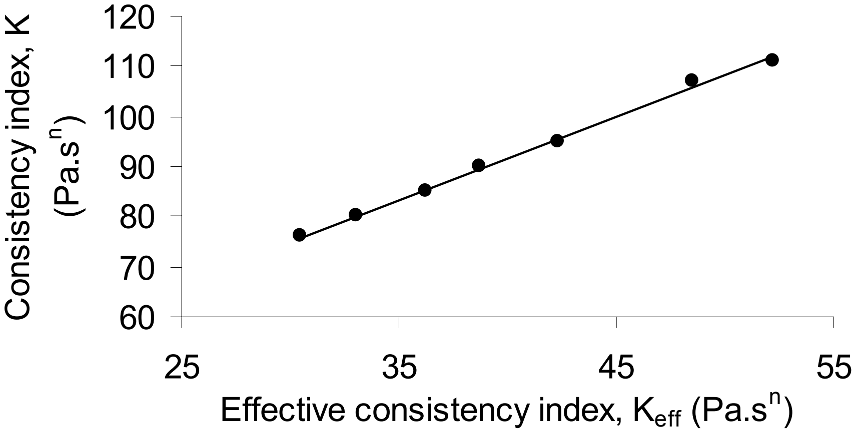

Fig. 8. Correlations developed between off-line consistency index K (rheometer) and Bostwick values with torque measurements for a series of tomato ketchup sample dilutions.

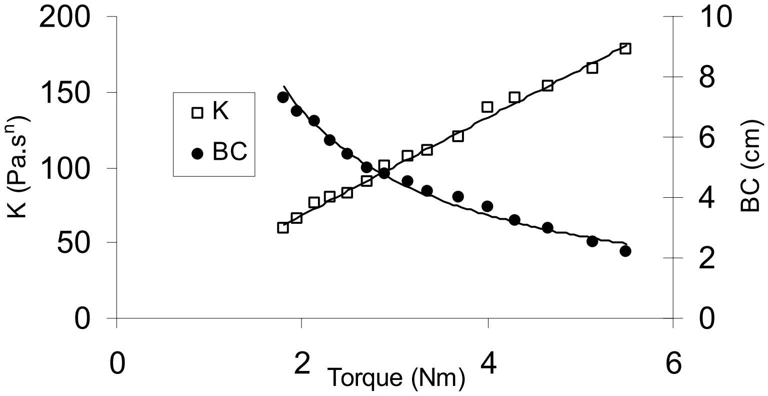

Table 1: Geometrical characteristics of helical ribbon agitator

Parameter	
Diameter, d (m)	0.36
Height, h (m)	0.31
Pitch, p (m)	0.18
Blade width, w (m)	0.05
Clearance, c (m)	0.01
p/d	0.5
c/d	0.027

Table 2: Flow characteristics of standard fluids, as determined by laboratory rheometer

Fluid	K	n
	Pa.s^n	-
Glycerol	5.55	0.98
Glycerol / H_2O / 0.1% CMC[a]	8.66	0.68
0.5% Carboxymethyl cellulose	10.3	0.56
1.0% Carboxymethyl cellulose	25.5	0.49
1.5% Carboxymethyl cellulose	65.6	0.43
0.5% Guar gum	25.4	0.33
1.0% Guar gum	80.9	0.25
1.5% Guar gum	253.3	0.16

[a] 85% mass glycerol, 15% mass water, 0.1% mass carboxymethylcellulose