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# Development and Parameter Estimation for a Multivariate Herschel-Bulkley Rheological Model of a Nanoparticle-Based Smart Drilling Fluid

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## Abstract

Smart drilling fluids containing Fe<sub>3</sub>O<sub>4</sub> nanoparticles have advantages toward increasing the hydraulic efficiency of drilling operations in a variety of reservoir environments. Exploring and optimizing the rheological behavior of such new drilling fluids is critical, implying direct and significant economic savings in developing new oil and gas fields. A experimental campaign analyzing the rheology of a bentonite-based fluid produced a new multiparametric dataset, considering a wide range of realistic reservoir conditions. Non-Newtonian behaviour is confirmed by yield stress computation for all these cases. Heating and rotation induce temperature and concentration gradients at drilling depth: it is hence essential to obtain an accurate but also versatile multivariate rheological model, which will enable viscosity prediction for the analyzed and other similar drilling fluids. The enhanced Herschel-Bulkley model is developed on a multiplicative assumption, postulating and analysing candidate equations which quantify the effect of shear rate, temperature and nanoparticle concentration on drilling fluid shear stress and viscosity. Parameter estimates have been subsequently determined via systematic optimisation, using statistical metrics to quantify and compare uncertainty and predictive potential. The trivariate shear stress and viscosity models proposed are similar in form: each requires six parameters used to combine a Herschel-Bulkley yield stress expression, an Arrhenius exponential of temperature and a linear model for nanoparticle concentration.

**Keywords:** parameter estimation, Herschel-Bulkley model, nanoparticles, drilling fluids.

## 1. Introduction

Efficient drilling for oil and gas requires the pumping of fluids with specific rheology, which can in principle be achieved by understanding the effect of composition on flow behavior and developing custom-made formulations with desired flow specifications; optimal drilling is thus attainable by matching rock, fluid and equipment characteristics. Flow behavior varies widely with concentration and temperature under constant shear, and successful tuning of rheological properties is critical to efficient drilling processes which achieve high penetration rates without risks of overheating, mechanical wear or excessive cost due to high fluid injection requirements (Sheng, 2011; Dandekar, 2013).

Developing correlations which express shear stress and viscosity as explicit functions of shear rate, temperature and concentration of iron nanoparticles (Waheed et al., 2014) is critical toward modeling, designing and planning of cost-effective drilling campaigns. Because of the multi-parametric uncertainty involved in rock strata mechanical properties, it is important to develop and validate high-fidelity predictive models which can provide online reliable estimates of rheological properties as explicit functions of environmental variables (temperature, mud concentration), to use equipment efficiently.

## 2. Experimental methods

Bentonite (Gold Seal, Halliburton) (tan powder, mild earthy odour, pH = 8-10) which consists of 1% cristobalite, 1% tridymite, 1-5% quartz and 60-100% actual bentonite and iron oxide nanoparticles (Sigma-Aldrich) in powder form (black powder, spherical shape, diameter of <50 nm, purity of > 97%) have been used to prepare all the samples. De-ionized water (TAMUQ, pH = 6.8-7.2) is used for base bentonite fluid preparation.

### 2.1. Sample preparation

Samples are prepared as per American Petroleum Institute (API) 13A-13B1 standards: 7% w/v bentonite dispersions of variable concentration in de-ionized water (600 ml) are obtained using a Hamilton Beach high-speed mixer for stirring (11,000 rpm for 20 min), and samples are left to hydrate and reach equilibrium in plastic containers (16 hr). Iron oxide nanoparticles (0.5, 1, 1.5, 2, 2.5, 3% v/v) are added slowly to avoid agglomeration. Before measuring rheological properties (by Couette viscometer–Brookfield rheometer), samples are mixed again (Hamilton Beach mixer, 5 min) to have identical shear history. This procedure is uniformly followed to ensure consistency and minimize bias effects.

### 2.2. Measurement procedures

A Couette (Grace M3600) viscometer and a vane (Brookfield YR-1) rheometer are used to measure rheological properties at three distinct temperatures and several nanoparticle concentrations (Yan & James, 1995); the Grace viscometer is equipped with a water bath and circulator to set the cup holder at the desired temperature (Fig. 1), so as to obtain and record shear stress and viscosity data (accuracy of experiments:  $T = \pm 2$  °C).

*Direct yield stress measurements* are obtained via the Brookfield vane rheometer using two different four-bladed vane spindles, to cover the extended yield stress range: the spindle is immersed in the test material and connected through a calibrated spiral spring to a motor drive shaft, rotating the vane at 0.1 rpm (optimal rheological analysis speed). Material resistance to movement is analysed via increasing torque values: shaft rotation is thus measured by means of calibrated spiral spring deflection via a rotary transducer.

*Indirect yield stress measurements* are obtained via the Grace rotational viscometer. Output parameters are shear rate ( $s^{-1}$ ), shear stress (Pa), viscosity (cP), gel strength (Pa). Viscometric data are obtained at fixed speeds (3, 6, 30, 60, 100, 200, 300, 600 rpm) and Newtonian shear rates (5.11, 10.21, 51.069, 102.14, 170.23, 340.46, 510.67, 1021.38  $s^{-1}$ ) are induced at the inner fixed cylinder, respectively (Kelessidis and Maglione, 2008). Readings are recorded every 10 s for a period of 60 s periods at each setting, giving 6 measurements at each of the 8 rotational speeds above (swept in descending order). Yield stress is estimated in all cases from viscometer rheograms, after extrapolating the shear stress-shear rate curve to zero shear rate and fitting the selected rheological model.

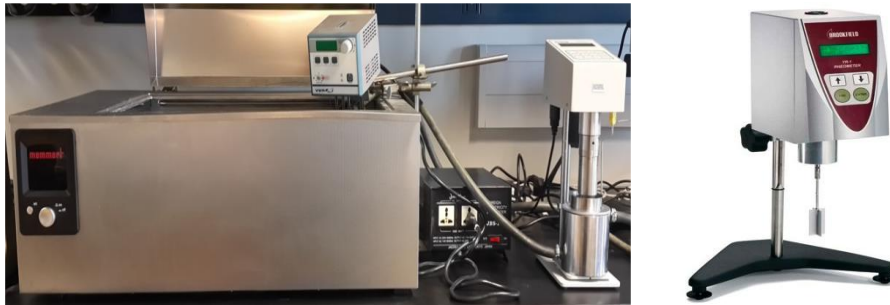


Figure 1: Grace M3600 viscometer with bath and circulator and Brookfield YR-1 rheometer.

### 3. Rheology modeling and parameter estimation

The prevalent correlation of shear stress and shear rate is the Herschel-Bulkley model (Kelessidis et al., 2006; Kelessidis & Maglione, 2008; Pouyafar & Sadough, 2013), a tri-parametric nonlinear correlation combining the effects of yield stress and power law behavior which outperforms the bi-parametric Bingham, power law and Casson models. The rheological parameters estimated from experimental campaigns are temperature- and concentration- dependent, and literature models are usually condition-dependent. Therefore, it is critical to first solve the optimisation problem for parameter estimation (both the univariate and the bivariate instance, for both shear stress and viscosity), and then develop a unified trivariate model, including all three independent variables (shear rate, temperature and iron nanoparticle concentration) in order to describe in detail the rheological behavior of the bentonite-based injection fluid during all drilling operations.

An extensive literature survey indicates the most widespread and suitable models of drilling fluids (Nguyen & Boger, 1992; Balmforth et al., 2014). Multivariate nonlinear least squares regression (Berge, 1993) has been employed in order to determine the most accurate model (Puxty et al., 2005) for shear stress and viscosity as functions of shear rate, temperature and additive concentration; the multi-parametric estimation problem has been subsequently solved using the novel experimental data, in order to compute the optimal set of parameters which minimise the sum of squared errors (SSE) and maximise the respective coefficient of determination,  $R^2$  (Graybill & Iyer, 1994). Ensuring that a global optimum is achieved is of particular importance, because strongly nonlinear multivariate expressions can induce solver trapping in various local minima; the multivariate nonlinear regression has thus been performed using a systematic strategy and several starting parameter sets (a multi-start strategy) to confirm optimality. Multivariate and multi-parametric nonlinear models are plotted vs experimental data, the standard error for shear stress and viscosity at each combination of independent variables has been computed, and error bars have been obtained to illustrate uncertainty.

Explicit multiparametric rheological models have the general form given in Eqs. (1)-(2). To simplify these we invoke the multiplicative assumption, illustrated in Eqs. (3)-(4). Univariate [Eqs. (5)-(6)], bivariate [Eqs. (7)-(10)] an trivariate [Eqs. (11)-(12)] models thus imply we can consider explicit correlations which are easy to validate accordingly.

$$\tau = f(\dot{\gamma}, T, C) \quad (1) \quad \mu = g(\dot{\gamma}, T, C) \quad (2)$$

$$\tau = f_1(\dot{\gamma})f_2(T)f_3(C) \quad (3) \quad \mu = g_1(\dot{\gamma})g_2(T)g_3(C) \quad (4)$$

$$\tau = k\dot{\gamma}^n + \tau_o \quad (\text{Herschel-Bulkley model}) \quad (5) \quad \mu = k\dot{\gamma}^n \quad (\text{Power law}) \quad (6)$$

$$\tau = (k\dot{\gamma}^n + \tau_o)(C + x) \quad (7) \quad \mu = k\dot{\gamma}^n(C + x) \quad (8)$$

$$\tau = (k\dot{\gamma}^n + \tau_o) \exp\left(-\frac{E}{RT}\right) \quad (9) \quad \mu = k\dot{\gamma}^n \exp\left(-\frac{E}{RT}\right) \quad (10)$$

$$\tau = (k\dot{\gamma}^n + \tau_o) \exp\left(-\frac{E}{RT}\right)(aC + b) \quad (11) \quad \mu = k\dot{\gamma}^n \exp\left(-\frac{E}{RT}\right)(C + b) \quad (12)$$

Figure 2: Explicit univariate and multivariate shear stress ( $\tau$ ) and viscosity ( $\mu$ ) parametric models.

## 4. Results and discussion

Nonlinear model plots have been obtained, confirming the rheological behavior of the samples is accurately described by an enhanced multivariate Herschel-Bulkley model for shear stress and a corresponding one for viscosity (Abu-Jdayil & Ghannam, 2014).

### 4.1. Univariate shear stress and viscosity models

Our shear stress and viscosity correlations for all temperatures appear in Fig. 3-4.

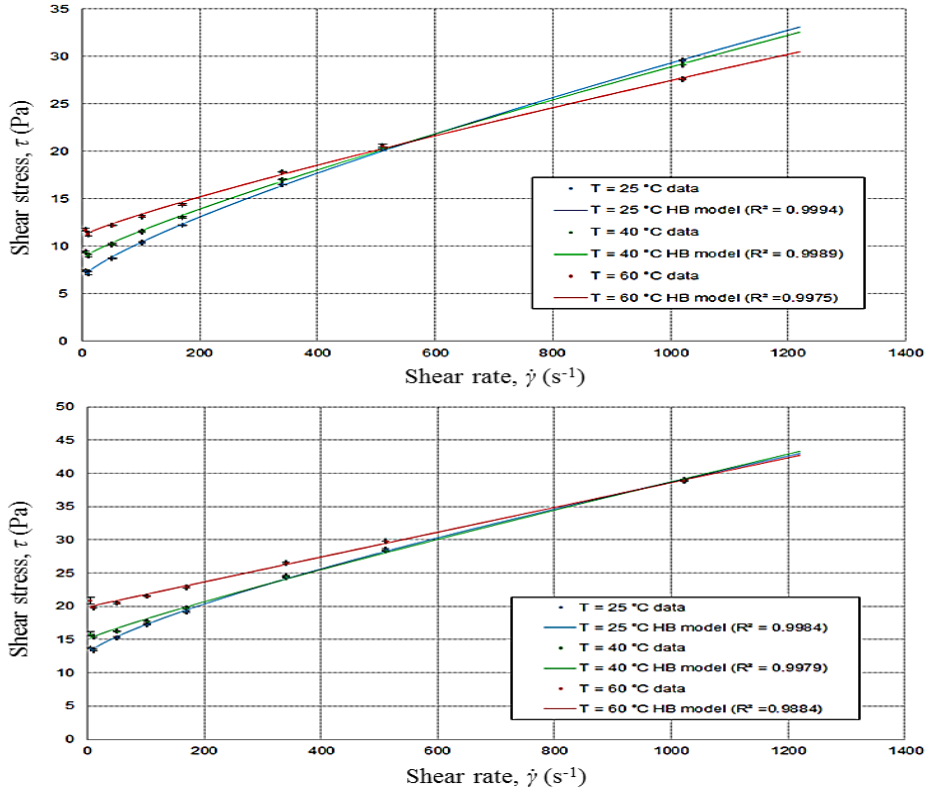


Figure 3: Shear stress vs. shear rate for all campaign temperatures (C = 0.5% up; C = 3%, down).

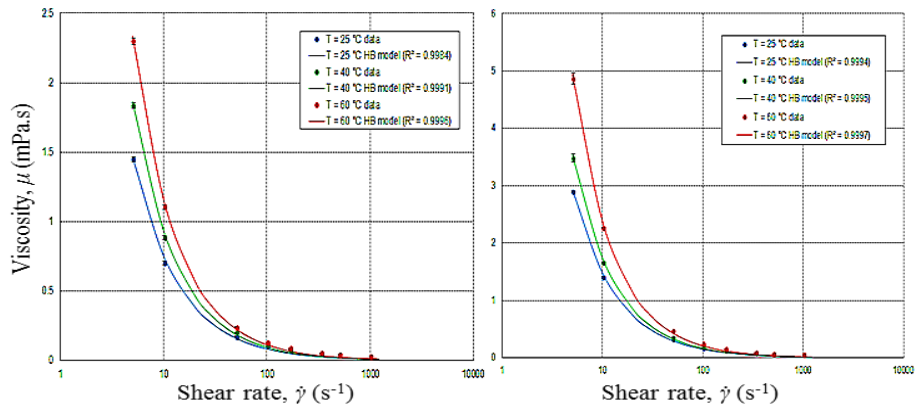


Figure 4: Viscosity vs. shear rate for all campaign temperatures (C = 0.5%, left; C = 3%, right).

#### 4.2. Bivariate shear stress and viscosity models

Our shear stress and viscosity correlations for all three temperatures appear in Fig. 5: to enhance clarity, viscosity surfaces have been separated by displacing the origin by  $\pm 2$ .

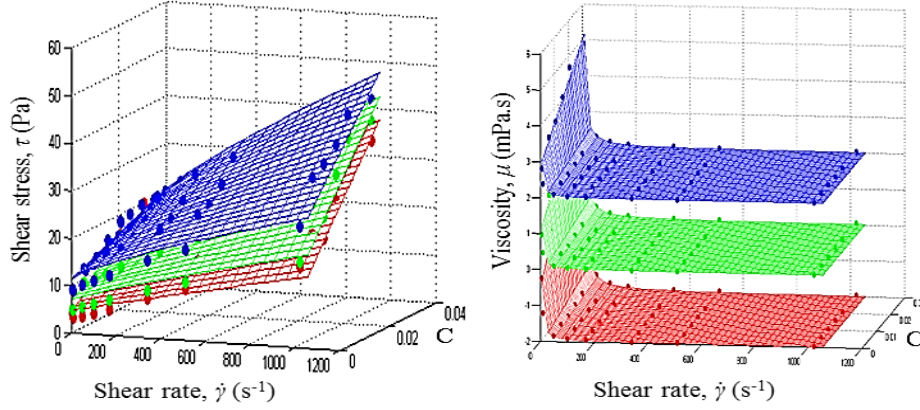


Figure 5: Shear stress (left) and viscosity (right) profiles vs. shear rate and vs. concentration.

Table 1: Parameter estimation for the univariate shear stress and viscosity parametric models.

C (%)	T (°C)	k (Pa.s <sup>-n</sup> )	n	τ <sub>0</sub> (Pa)	R <sup>2</sup>	SSE	k (kgm <sup>-n</sup> )	n	R <sup>2</sup>	SSE
0	25	0.09686	0.7471	3.4915	0.9983	0.39	3.7478	-0.9486	0.9950	0.0027
	40	0.03268	0.8676	4.6598	0.9995	0.08	4.6289	-0.9647	0.9987	0.0011
	60	0.01959	0.9448	8.2149	0.9934	1.02	10.2237	-1.0810	0.9985	0.0040
0.5	25	0.10025	0.7843	6.7007	0.9994	0.25	6.84891	-0.9588	0.9984	0.0041
	40	0.05953	0.8432	8.7383	0.9989	0.38	9.23222	-0.9946	0.9991	0.0037
	60	0.04329	0.8591	11.103	0.9975	0.56	11.8929	-1.0110	0.9996	0.0030
1.0	25	0.07794	0.8216	7.9654	0.9997	0.13	7.78388	-0.9521	0.9991	0.0031
	40	0.03467	0.8886	8.6692	0.9981	0.42	9.41873	-0.8595	0.9993	0.0267
	60	0.02751	0.9011	11.2771	0.9956	0.73	12.2908	-1.0225	0.9997	0.0025
1.5	25	0.09331	0.8015	9.8931	0.9988	0.56	10.4552	-0.9895	0.9990	0.0055
	40	0.06237	0.8371	12.2577	0.9983	0.59	12.9174	-1.0002	0.9995	0.0039
	60	0.01869	0.9704	16.8437	0.9940	1.18	18.7670	-1.0383	0.9997	0.0045
2.0	25	0.12702	0.7696	12.8649	0.9991	0.50	13.1417	-0.9765	0.9994	0.0057
	40	0.05029	0.8910	15.0585	0.9977	1.10	15.8788	-1.0024	0.9997	0.0041
	60	0.01907	0.9971	19.9311	0.9957	1.29	22.1101	-1.0376	0.9997	0.0064
3.0	25	0.10828	0.7910	13.7773	0.9984	0.87	14.5247	-0.0993	0.9994	0.0064
	40	0.03905	0.9236	16.8603	0.9979	0.95	18.4641	-1.0270	0.9995	0.0075
	60	0.01182	1.0563	23.4468	0.9884	3.05	27.2611	-1.0601	0.9997	0.0102

Table 2: Parameter estimation for the bivariate shear stress and viscosity parametric models.

C (%)	k (Pa.s <sup>-n</sup> )	n	τ <sub>0</sub> (Pa)	E/R (K)	R <sup>2</sup>	SSE	k (kg.m <sup>-n</sup> )	n	E/R (K)	R <sup>2</sup>	SSE
0	0.2721	0.8528	38.69	608.7	0.9296	41.22	10880	-1.035	2364	0.992	0.0334
0.5	0.1225	0.8263	16.72	198.4	0.9707	29.40	470.8	-0.995	1238	0.998	0.0130
1.0	0.2335	0.7122	16.92	214.2	0.9732	22.17	209.7	-1.003	965.1	0.997	0.0264
1.5	0.1758	0.8583	44.54	384.8	0.9436	60.05	1206	-1.017	1407	0.998	0.0221
2.0	0.1647	0.8783	50.76	361.8	0.9613	53.64	697.6	-1.013	1171	0.998	0.0360
3.0	0.2521	0.915	118.8	590	0.9411	83.29	2068	-1.037	1464	0.998	0.0567

T (°C)	k (Pa.s <sup>-n</sup> )	n	τ <sub>0</sub> (Pa)	b	R <sup>2</sup>	SSE	k (kg.m <sup>-n</sup> )	n	b	R <sup>2</sup>	SSE
25	2.262	0.7882	221.1	0.02921	0.9600	1.43	346.1	-0.980	0.01408	0.9889	0.26
40	1.168	0.8823	311.4	0.02297	0.9399	1.94	429.7	-1.010	0.01421	0.9888	0.37
60	0.4955	0.9726	419.5	0.02351	0.9482	1.47	594.7	-1.044	0.01547	0.9936	0.39

## 5. Conclusions

Our development of an enhanced multivariate Herschel-Bulkley rheological model for a bentonite-based candidate drilling fluid with variable Fe<sub>3</sub>O<sub>4</sub> nanoparticle composition is based on original data from an experimental campaign in a wide range of conditions, followed by comparative evaluation of multivariate shear stress and viscosity models. Parameter estimation via systematic optimization and comparative statistical analysis of the resulting uncertainty metrics allows the assessment of candidate expressions toward determining their suitability and quantifying their reliability for modeling and design. The shear stress and viscosity correlations describe the rheological effects of shear rate, temperature and Fe<sub>3</sub>O<sub>4</sub> nanoparticle concentration: they encompass a Herschel-Bulkley model, an Arrhenius temperature exponential and a linear equation, respectively, achieving a high predictive potential in both bivariate ( $R^2_{\tau(\gamma,T)} = 0.929$ ,  $R^2_{\mu(\gamma,T)} = 0.992$ ) as well as in the full trivariate version of the model ( $R^2_{\tau(\gamma,T,C)} = 0.987$ ,  $R^2_{\mu(\gamma,T,C)} = 0.988$ ). Higher temperatures induce increased standard error for both shear stress and viscosity; heating affects fluid structure and rheology (hence prediction uncertainty) appreciably. Further investigation of concentration effects for various additives is planned, but the models already provide reliable estimates in a wide range of realistic drilling conditions.

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