Rheological behavior of aqueous suspensions of gray clay and modeling their transport in flexible pipes

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Abstract:
This work is divided into two distinct but complementary parts. The first part deals with the study of the effect of concentration on the rheology of aqueous solutions of gray clay. Of particular interest is the case where the concentration is 50%, since it has been necessary to insert a specific approach for the determination of the rheological parameters introduced by the Robertson-Stiff model law of behavior. In the second part, we study the general case of the flow of a fluid described by this model in flexible conduct and the purpose of which is to determine “flow-pressure drop” type relationships that could allow more control of these flows.

Keywords:
Rheology/Gray clay/flexible pipes/Robertson-Stiff/flow/pression

1 Introduction
As is the case with composite materials, clay-based materials continue to play a very important role in various areas of modern industry; this is particularly the case in civil engineering, in the cosmetics industry or for the pharmaceutical industry [1]. The development of these materials is most often declined through the mixture of a sifted clay powder and some often polymeric additives with distilled water thus allowing the formation of pasty solutions that evolve to a more or less orderly form; the speed or the control of this evolution remaining dependent on the physicochemical properties of these solutions or the conditions of their elaboration. Also and in a logic of quality control, tests are often conducted at different levels to ensure that the operating conditions previously agreed are taken into account; these frequently relate to the mechanical and / or thermal properties of the finished product or to properties related to the transport of these products in the fresh state in a confined environment. In this context and whatever the purpose sought, the characterization of the rheological behavior of aqueous solutions based on clay today has a certain importance in terms of prediction and / or monitoring for the manufacture of these materials.

Our work is part of this approach and is divided into two complementary parts; a purely experimental first part where we describe, using the Rheostress I rotary rheometer (controlled by Rheowin software), the effects of the formulation of aqueous solutions of a gray clay taken from the Moroccan atlas on their rheological behavior and a purely theoretical second part where we propose a pressure drop-flow relationship when it is assumed that these aqueous solutions are transported in flexible pipes.

For the first part, we show in particular that the variation of the clay powder concentration makes it possible to identify several forms of shear thinning behavior with or without threshold. Particular interest is given to the case where the mass of the liquid phase is twice as large as that of the solid phase insofar as the rheological models listed on the Rheowin software do not allow to approach
the experimental readings with a good regression. The use of an error optimization method based on the calculation of standard deviations enabled us to verify for this particular case that the rheological behavior is very close to that observed by Robertson and Stiff in 1976 for the case of sludge drilling [2].

In the second part of this work, we propose, for the control of the transport of these solutions in flexible pipes, a set of approximate relations making it possible to connect the transported flow with the pressure drop imposed. In this context we insist on the particular case where the fluid is described by the Robertson Stiff model on the one hand and where the transmural pressure is connected to the duct section by a constitutive equation similar to that proposed by Rumberger and Nerem in 1977 on the other hand. The method used is based on an asymptotic treatment of the conservation equations integrated on a section in the case where the number of Womersley is supposed to be very weak in front of the unit and where the terms of parietal friction are determined from a corrected form of the expression of Poiseuille.

2 Impact of the concentration on the rheological behavior of aqueous suspensions of gray clay:

2.1 Raw materials and elaboration

The gray clay that we consider here comes from the Middle Atlas of Morocco. This clay is poor in carbonate (CaO + MgO), it is mainly composed of SiO₂ silica (58.06%) and contains a significant percentage of K₂O. The spectrum obtained from an X-ray diffraction test shows the presence of free silica in the form of quartz SiO₂, muscovite KAl₃Si₃O₁₀(OH)₂ and halloysite Al₂Si₂O₅(OH)₄·2H₂O as clay mineral phase (fig.1).

![Figure 1: DRX of gray Clay](image)

For the development of the samples that are the subject of this study, we proceed as follows:
• The sieving of the clay powder (the scale of the orifices used is 500 μm)
• The addition of distilled water according to the desired concentrations (the ratios [dry matter (S) / liquid matter (L)] retained are 25%, 50%, 75% and 100%)
• Mixing through an agitator containing a magnetic bar with a speed of 6 r / s for 5 min.

2.2 Test bench and measurement protocol:

The rotary rheometer used is a Haake Rheostress1 from "Thermo Scientific" operating at an imposed speed and controlled by Rheowin software. The tests are carried out in cone-plane cell (CP60) for which the lower plane is fixed and the cone is driven in a rotational movement about their
common axis; these components are sealed in Titanium and their geometry is characterized by their diameter (60 mm), their hardness coefficient (6), their air gap (1 mm) and their truncation angle (α = 1°).

The imposed rotation signal is triangular: it is characterized by its rise time $T_0$ and its maximum rotation $\Omega_{\text{max}}$ (fig. 2). The values that we use for these variables are those that minimize the effects due to the inertia of the device [3], in this case the measurements of the shear stress as a function of the corresponding shear rate $\dot{\gamma}$ in the ascending phase and the downward phase are almost confused and the configuration of the test is quasi-stationary.

\[
\begin{align*}
\Omega(t) &= \frac{\Omega_{\text{max}}}{T_0} t & \text{for} & & 0 \leq t \leq T_0 \\
\Omega(t) &= \Omega_{\text{max}} \left(2 - \frac{t}{T_0}\right) & \text{for} & & T_0 \leq t \leq 2T_0
\end{align*}
\]  

(1)

![Figure 2: Rheological test imposed](image)

3 study of the rheological behavior of aqueous suspensions of gray clay:

The aim here is to highlight the effects due to the variations of the concentration on the rheology of the solutions through the determination of the optimal law of behavior corresponding to each of the values of $R$ retained in this work.

3.1 Results and discussion

In the following table, we present, for these concentrations, a synthesis relating to the correlation of the rheological behavior directly deduced from the operation of the Rheowin software.

<table>
<thead>
<tr>
<th>Concentration</th>
<th>Rheological behavior (Rheowin)</th>
<th>Coefficient of 'determination</th>
<th>Rheological parameters</th>
</tr>
</thead>
<tbody>
<tr>
<td>25%</td>
<td>Newton</td>
<td>0.9875</td>
<td>$\eta = 0.002369 \text{ Pa.s}$</td>
</tr>
<tr>
<td>50%</td>
<td>Bingham, Ostwald Waele, Herschel-Bulkley</td>
<td>0.3559, 0.2448, 0.3089</td>
<td></td>
</tr>
<tr>
<td>75%</td>
<td>Bingham</td>
<td>0.9863</td>
<td>$\tau_0 = 0.02189 \text{ Pa}$, $\eta_p = 0.005830 \text{ Pa.s}$</td>
</tr>
<tr>
<td>100%</td>
<td>Ostwald Waele</td>
<td>0.9850</td>
<td>$K = 0.03827 \text{ Pa.s}^n$, $n = 0.778$</td>
</tr>
</tbody>
</table>

It shows that apart from the case where $R = 50\%$, the different behaviors displayed for $R = 25\%, 75\%$ and $100\%$ correspond to regression factors between the experimental survey and the model listed on the software of the order of 0.98. For $R = 50\%$ (fig. 3), the regression factors displayed are less than 0.35, which indicates a poor description of the experimental record by these laws. (Tab. 1)
Also and in the light of the scientific reflection on this problem [2,4], we can notice that several forms of generalization for the Bingham and Ostwald Waele models, other than that of Herschel Bulkley, are often proposed to correct differences observed in this case. [1]

In this context, we chose to retain the model introduced by Robertson and Stiff in 1976 to describe the rheological behavior corresponding to R = 50% [1, 4].

For a shear test, this behavior is described by:

\[
\begin{align*}
\tau > \tau_0: & \quad \tau = A(B + \dot{\gamma})^C \\
\tau < \tau_0: & \quad \dot{\gamma} = 0
\end{align*}
\]  

It is a threshold fluid described by a three-parameter model (A, B and C) from which we can find the model of Bingham (C = 1) and that of Ostwald Waele (B = 0) and for which the threshold stress (\(\tau_0\)), the consistency (k) and the melt index (n) are respectively defined by:

\[
\tau_0 = A B^n, \quad k = A, \quad n = C, \quad (3)
\]

From a practical point of view and for the determination of these parameters, we implement the following algorithm using version 3.6.7 of the Python software:
For the particular case that concerns us here (R = 50%), the quality of the regression results displayed (fig. 4) thus makes it possible to affirm that the rheological behavior of the solution is described by the shear thinning threshold model \( \tau = 0.15(1.6 + \dot{\gamma})^{0.4} \) for \( \dot{\gamma} > 0.18 \).

For the validation of this approach, we have reconsidered the case treated in the literature [5] for a comparison between the results obtained from the execution of the algorithm above and those displayed from the method "Golden section search method". We observe (fig. 5) an almost perfect agreement between the plots corresponding to each of the two methods.

<table>
<thead>
<tr>
<th>Table 2: Gap between our results and [5]</th>
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<tbody>
<tr>
<td></td>
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<tr>
<td>( \tau_0 (Pa) )</td>
</tr>
<tr>
<td>( K (Pa.s^n) )</td>
</tr>
<tr>
<td>( n )</td>
</tr>
</tbody>
</table>

Figure 4: Fit curve with the values of the parameters

Figure 5: Validation of the results obtained by the Golden section search method
4 Modeling the flow of gray clay suspensions in flexible pipe:

In this second part we propose, for the control of the transport of these solutions in flexible pipes, approximate relations making it possible to connect the transported flow with the pressure drop imposed. The approach used is based on the introduction of a tube law and an asymptotic treatment of integrated conservation equations on a section.

4.1 Mathematical formulation:

In the system of cylindrical coordinates, we consider the unsteady flow of an incompressible threshold fluid in an elastic, axisymmetric, impermeable pipe of length $L$, of mean radius $R_0$, of axis of revolution $e_z$.

![Figure 6: Geometry of the flow](image)

The velocity field of the fluid particles is defined by:

$$\overrightarrow{V}(M) = V_r (r, z, t) \overrightarrow{e_r} + V_z (r, z, t) \overrightarrow{e_z}$$  \hspace{1cm} (4)

and the boundary conditions are those that translate:

a- The symmetry with respect to the axis of the duct:

$$\begin{bmatrix}
V_r (0, z, t) = 0 \\
\tau_{rz} (0, z, t) = 0
\end{bmatrix}$$  \hspace{1cm} (5)

b- The adhesion of the fluid to the wall:

$$\begin{bmatrix}
V_r (R, z, t) = \frac{\partial R (z, t)}{\partial z} \\
V_z (R, z, t) = 0
\end{bmatrix}$$  \hspace{1cm} (6)

We place ourselves more in the case where:

- The flow is unidirectional (the parameter of form $\varepsilon = \frac{R_0}{L_0}$ very weak in front of the unit).
- Voluminal forces are neglected.
- The pressure forces are comparable to the viscous forces.
- The evolution of the section as a function of the transmural pressure is given by:

$$A(z, t) = A_0 (z) \ e^{\frac{P(z,t)-P_{ext}}{\xi}}$$  \hspace{1cm} (7)

Or:

$P_{ext}$ is the pressure exerted by the external environment on the wall

$P (z, t)$ is the pressure at a point of the fluid

$A_0$ is the section at zero transmural pressure
ξ is a constant characterizing the rigidity of the wall.

This law, already used by other authors [6], allows to find the case of a linear evolution of the pressure according to the section considered [7], it also allows to integrate, if necessary, evolutions non-linear of these variations.

\[ \frac{\partial Q(z,t)}{\partial t} + \frac{\partial}{\partial z} \left( \alpha \frac{Q(z,t)^2}{A(z,t)} \right) = \left( - \frac{\partial p(z,t)}{\partial z} \right) A(z,t) + F(z,t) \] (8)

Where:
\[ \alpha(z,t) = \frac{2\pi A(z,t)}{Q(z,t)^2} \int_0^{R(z,t)} \left( \frac{Q(z,t)}{Q(z,t)^2} \right) r \, dr \] (9)

And:
\[ F(z,t) = \frac{2}{\rho} \sqrt{\pi A(z,t)} \tau_p \] (10)

It is therefore a system of partial nonlinear differential equations where the unknowns are (Q, P) and where it is common to introduce additional simplifying hypotheses [9] for the approximation of the correction factor α(z, t) and the parietal friction force F(z, t). Note that in the general case, the exact determination of these quantities remains dependent on the resolution of the system of conservation equations in local formulation.

4.2 Resolution procedure:

For the resolution of the system (8.9.10) we retain the following approximations [9]:

- \[ \alpha = 4/3 \]

- \[ F(z,t) = \frac{dp}{dz} \frac{A(z)}{\rho} \] (11)

This approximation of F(z, t) is deduced from the resolution of the system of local equations in the case where the pressure depends only on z and where the inertial forces are negligible compared to the viscous forces.
4.2.1 Determination of an approximation of $F(z, t)$ for the particular case of a threshold fluid described by the Robertson-Stiff model:

Taking into account the hypotheses introduced above and the constitutive law presented in (2), one can obtain a first approximation of the flow from the local system, this one is given by:

$$Q = \left[ \frac{1}{2k} \right]^\frac{1}{\zeta} \frac{1}{\pi} \left( \frac{3}{2} \right)^\frac{1}{\zeta} \left( \frac{A(z)}{\frac{1}{2C}} + \frac{1}{\frac{1}{2C}} \right) \left[ -\frac{dp}{dz} \right]^{\frac{1}{\zeta}} A(z)^{\frac{1+3C}{2C}}$$

And was deduced from the assumption [10]:

$$\lambda = \frac{r_0}{R(z,t)} = \frac{\tau_s}{\tau_{p(z,t)}} = \text{cst}$$

Where $r_0$ is the radius of the plug zone.

By eliminating $dp/\,dz$ between (1) and (2) the expression of $F(z)$ that we consider in the rest of this work:

$$F(z) = -k \frac{Q^C}{A(z)^{\frac{3C-1}{2}}} \quad \text{with} \quad k = \frac{2K(3C + 1)^\frac{1+3C}{2}}{\rho C^\frac{3}{2} \left( \frac{3}{2} \right) \left( \frac{1}{2C} + \frac{1}{C} \right)^{1+3C}}$$

We find from (14) classical expressions introduced in previous works [9-10] where the models of behavior retained are either non-Newtonian with yield stress (Bingham) ($n=1$) or without yield stress (Ostwald Waele) ($\tau_0=0$).

4.2.2 Treatment of the global system:

Under these conditions, the quasi-stationary form of the system (8) gives:

$$\left\{ \begin{array}{l}
\frac{\partial Q}{\partial z} = 0 \\
\frac{A(z)}{\partial z} 
\end{array} \right\}$$

The processing of the system (15) thus provides access to the implicit equation (16) connecting the flow rate to the pressure drop.

$$Q^C = \frac{\alpha}{kL} \left( \frac{3(C-1)}{2} - \left( A_0 \frac{P_{in} - P_{out}}{\xi} \right) \frac{3(C-1)}{2} \right) \frac{Q^2}{\rho kL} \left( \frac{3c+1}{2} - \left( A_0 \frac{P_{in} - P_{out}}{\xi} \right) \frac{3c+1}{2} \right) = 0$$

It should be noted that compared to the previous works, an additional correction in the expression of the flow is introduced, it is declined through the taking into account of the convective terms in (15).
It can be seen that the evolution of the flow as a function of pressure is non-linear. When the pressure drop increases, the flow rate increases. This evolution is due to the non-linearity of the convective terms, the tube law, the elasticity of the wall and the law of behavior.

It should be noted that the rheological parameters strongly affect the evolution of the flow rate as a function of the pressure drop. The increase in consistency (A) and melt index (C) imply a decrease in flow rate. The control of the flow of a Robertson-Stiff fluid is based on the monitoring of its rheological parameters.

5. Conclusion:

In this work, we present the impact of the clay powder concentration on the rheological behavior of aqueous solutions. We have previously identified the rheological nature of the solutions considered. For a concentration of 50% in gray clay, its behavior does not correspond to the models listed on the Rheowin. The use of an adjustment and regression method allowed us to verify for this case that the rheological behavior is described by Robertson and Stiff model.

The control of the flow in flexible pipe for this type of fluid was the subject of a second part allowing the theoretical determination of the evolution of the flow as a function of the pressure drop,
for this evolution, the nonlinear effects due to the fluid behavior and the tube’s deformability are considered. This consolidates flow control processes from rheological nature on the one hand and hydrodynamic parameters on the other.

Acknowledgements


References