Investigation of feasibility of a new method to characterise porous media in terms of pore size distribution

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1 Introduction

Current methods employed to determine pore size distributions in porous media present several drawbacks such as their toxicity (mercury porosimetry). An original method to measure pore size distribution using yield stress fluids has been proposed in the literature [1],[2]. The main idea in these works is that using fluids with a threshold below which the fluid does not flow allows obtaining the pore size distribution by measuring the evolution of the flow rate versus pressure gradient. These authors have solved the inverse problem giving the pore size distribution for Bingham, Herschel-Bulkley (H-B) and Casson fluids. The objective of our work is to investigate the experimental feasibility of the method proposed. In this paper, relationships between the properties of the pump and those of the yield stress fluid are proposed allowing the measurement of the pore-size distribution of a model porous medium.

2 Laminar flow through capillaries for Herschel-Bulkley fluids

The rheological law that rules the behaviour of H-B fluids is given by [3]:

$$\tau = \tau_0 + k\dot{\gamma}^n \tag{1}$$

where τ and τ_0 are respectively the shear stress and the yield stress below which the fluid does not flow. $\dot{\gamma}$, k and n are the shear rate, consistency and flow behaviour index respectively.

As proposed in [1], the porous medium is idealized as a bundle of capillaries with a given radius distribution. Since a flow through a capillary is supposed, the shear rate is given as the radial derivative of the velocity ($\dot{\gamma} = (\partial u/\partial r)$). The yield stress is then associated to a critical radius given by:

$$r_0 = \frac{2\tau_0}{\nabla P} \tag{2}$$

where ∇P is the pressure gradient between the bounds of the capillary tube. For r<r_0 velocity profile is uniform. That means that the fluid does not flow through the pores with a radius smaller than r_0 because of no-slip boundary condition. The flow rate of a H-B fluid through a capillary of radius r on which a pressure gradient ∇P is imposed can be written as [4]:

$$q(\nabla P, r) = \left[1 - 2\left[\frac{\left(1-\frac{2\tau_0}{\nabla Pr}\right)^2}{\left(\frac{1}{n}\right)+3} + \frac{\frac{2\tau_0}{\nabla Pr}\left(1-\frac{2\tau_0}{\nabla Pr}\right)}{\left(\frac{1}{n}\right)+2}\right]\right]\left[\frac{r}{\left(\frac{1}{n}\right)+1}\left(\frac{r\nabla P}{2K}\right)^{\left(\frac{1}{n}\right)}\left(1-\frac{2\tau_0}{\nabla Pr}\right)^{\left(\frac{1}{n}\right)+1}\right]\pi r^2 \tag{3}$$

3 Characterising porous media samples in terms of pore-size distribution

Considering a porous medium with a pore size probability density function p(r) in frequency, the average flow rate per pore is given by,

$$\overline{q(\nabla P)} = \frac{Q(\nabla P)}{N} = \int_0^\infty q(\nabla P, r)p(r)dr \tag{4}$$

Where $N = R^2\emptyset/\bar{r}^2$, $\emptyset$ is the porosity, R is the sample radius and $\bar{r}$ is the average pore size in the sample. Using equations (3) and (4), and the derivation rule

$$\frac{\partial}{\partial \nabla P}\int_{r_0(\nabla P)}^\infty f(\nabla P, r)dr = \int_{r_0(\nabla P)}^\infty \frac{\partial}{\partial \nabla P}[f(\nabla P, r)]dr - f(\nabla P, r_0)\frac{\partial}{\partial \nabla P}r_0, \tag{5}$$

and keeping in mind the fact that the fluid does not flow in capillaries with r<r_0, the inversion of equation (4) leads to the following expression for the probability density function for any integer value of 1/n:

$$p(r_0) = [(\frac{1}{n}+4)\frac{\partial^{\frac{1}{n}+1}\overline{q(\nabla P)}}{\partial \nabla P^{\frac{1}{n}+1}} + \nabla P\frac{\partial^{\frac{1}{n}+2}\overline{q(\nabla P)}}{\partial \nabla P^{\frac{1}{n}+2}}]\frac{2^{\frac{1}{n}+4}K^{\frac{1}{n}}\nabla P}{16\,(\frac{1}{n})!\pi r_0^{\frac{1}{n}+4}} \tag{6}$$

This last relation is valid for all values of ∇P, that is for all values of r_0. That means that a linear combination of two derivatives of the average flow rate per pore relative to ∇P would allow obtaining the pore size distribution.

4 Sizing the experiment

The experimental set up which is investigated in this paper is, *a priori*, a classical one: the fluid is injected in a sample of porous medium using a pump and the pressure loss over the sample is recorded for a range values of imposed fluid flow rate. In order to size the experiment, the knowledge of the pore radius sizes with a cumulated probability between 5% and

95% is considered sufficient. Hence, pore radii whose cumulated probabilities are 5% and 95% are denoted r_{min} and r_{max} respectively. Moreover, $r_{final} = 2r_{max}$ is considered to be the biggest radius (cumulated probability close to 100%).

To calculate the probability p(r) of a determined pore size, r, all that is needed is to substitute ∇P by $2\tau_0/r$ in Eq. (6), thus making this radius a critical one. r_{min} determines the maximum pressure gradient (and then the maximum flow) and r_{max} the minimum one. The minimum and maximum flow rates, $Q_{min}(\nabla P_{min})$ and $Q_{max}(\nabla P_{max})$, to be imposed by the pump are respectively the ones to penetrate 5% (the largest pore-sizes) and 95% of the pores of the sample. These quantities can then be numerically determined by introducing (2) and (3) into (4):

$$Q_{min} = \frac{R^2 \emptyset}{\bar{r}^2} \sum_{r_{max}}^{r_{final}} \left[1 - 2\left[\frac{\left(1-\frac{r_{max}}{r}\right)^2}{1/n+3} + \frac{\frac{r_{max}}{r}\left(1-\frac{r_{max}}{r}\right)}{1/n+2}\right]\right] [\frac{r}{\frac{1}{n}+1} \left(\frac{r\tau_0}{kr_{max}}\right)^{\frac{1}{n}} \left(1-\frac{r_{max}}{r}\right)^{1+\frac{1}{n}}]\pi r^2 p(r) \quad (7)$$

$$Q_{max} = \frac{R^2 \emptyset}{\bar{r}^2} \sum_{r_{min}}^{r_{final}} \left[1 - 2\left[\frac{\left(1-\frac{r_{min}}{r}\right)^2}{1/n+3} + \frac{\frac{r_{min}}{r}\left(1-\frac{r_{min}}{r}\right)}{1/n+2}\right]\right] [\frac{r}{\frac{1}{n}+1} \left(\frac{r\tau_0}{kr_{min}}\right)^{\frac{1}{n}} \left(1-\frac{r_{min}}{r}\right)^{1+\frac{1}{n}}]\pi r^2 p(r) \quad (8)$$

It is accepted that the experimental restrictions are in terms of ΔP_{max}, ΔP_{min}, Q_{max}, Q_{min} (choice of the pump) and in terms of τ_0, n and k (choice of the fluid). For a given range of main pore-sizes (that is pore-size with a cumulated probability between 5% and 95%), imposing the choice of the pump leads to the properties of the fluid needed for the experiments. By rewriting (2), (7) and (8), the following inequalities allowing the sizing of the experiment are obtained:

$$\frac{r_{max}}{2L}\frac{\Delta P_{min}}{L} \leq \tau_0 \leq \frac{r_{min}}{2L}\frac{\Delta P_{max}}{L} \quad (9)$$

$$k \geq \frac{\tau_0}{Q_{max}{}^n} [\left(\frac{1}{r_{min}}\right)^{\frac{1}{n}} \frac{R^2\emptyset}{\bar{r}^2} \sum_{r_{min}}^{r_{final}} \left[1 - 2\left[\frac{\left(1-\frac{r_{min}}{r}\right)^2}{\frac{1}{n}+3} + \frac{\frac{r_{min}}{r}\left(1-\frac{r_{min}}{r}\right)}{\frac{1}{n}+2}\right]\right] \left[\frac{r}{\frac{1}{n}+1}\left(1-\frac{r_{min}}{r}\right)^{1+\frac{1}{n}}\right] \pi r^{2+\frac{1}{n}} p(r)]^n \quad (10)$$

$$k \leq \frac{\tau_0}{Q_{min}{}^n} [\left(\frac{1}{r_{max}}\right)^{\frac{1}{n}} \frac{R^2\emptyset}{\bar{r}^2} \sum_{r_{max}}^{r_{final}} \left[1 - 2\left[\frac{\left(1-\frac{r_{max}}{r}\right)^2}{\frac{1}{n}+3} + \frac{\frac{r_{max}}{r}\left(1-\frac{r_{max}}{r}\right)}{\frac{1}{n}+2}\right]\right] \left[\frac{r}{\frac{1}{n}+1}\left(1-\frac{r_{max}}{r}\right)^{1+\frac{1}{n}}\right] \pi r^{2+\frac{1}{n}} p(r)]^n \quad (11)$$

Note that in the last two inequalities, the pore size distribution p(r) is *a priori* unknown. So these are useful only to size an experimental system devoted to a given range of porous media in terms of pore distribution size distribution. In order to illustrate this last remark, a normal pore size distribution is now considered. Its average radius is related to the intrinsic porosity of a model porous medium sample of a sintered silicate:

$$\bar{r} = \sqrt{(8K/\emptyset)} \quad (12)$$

where K is the permeability. For such a medium, typical values of intrinsic permeability and porosity are K=8 $10^{-12}m^2$ and $\emptyset$=0.45. The pores sizes are distributed according to a normal distribution of average 12µm, standard deviation 3µm and r_{final}=24µm as shown in Figure (1). Considering a cylindrical sample of radius R=2.5cm and of length L=0.1m, and assuming that the pump (Teledyne ISCO 500D[5])is able to provide [$Q_{min}=1.67*10^{-11}m^3/s$; $Q_{max}= 3.4*10^{-6}m^3/s$; ΔP_{min}=0.69bars; ΔP_{max}=258.75bars], a H-B fluid with a flow behaviour index n=0.5 has to satisfy the following constraints for its yield stress and its consistency: $5.840Pa \leq \tau_0 \leq 914.086Pa$ and $\tau_0/40.725 \leq k \leq \tau_0/3.557$. Alumina nanoparticles suspensions may respect theses conditions[6].

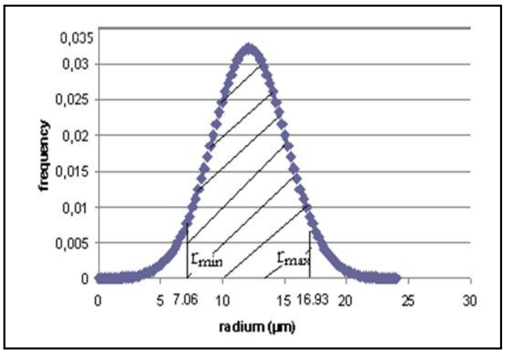

Figure 1 – Sample with a Normal distribution of average 12µm and standard deviation 3µm

Conclusion

A method has been proposed to size an experimental setup of H-B fluid injection in a porous medium. These preliminary calculations show that such a setup can probably be considered at least to characterise the pore size distribution of a classical sintered silicate. The aim is now to build this setup and test it.

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