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Notes based on:

# Experimental and Theoretical Memory Diffusion of Water in SAND

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#### NOTES BASED ON

### EXPERIMENTAL AND THEORETICAL MEMORY DIFFUSION OF WATER IN SAND. (Submitted for publication)

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#### Abstract

The basic equations used to study the fluid diffusion in porous media have been set by Fick and Darcy in the mid of the XIXth century, although earlier all these equations were imbedded in the classic Fourier equation of diffusion. However some data on the flow of fluids in rocks exhibit properties which may not be interpreted with the classical theory of propagation of pressure and fluids in porous media [Bell and Nur, 1978; Roeloffs, 1988] and other phenomena, as propagation of tides of the ocean in subterranean waters (Robinson 1939) have non yet found a mathematical model . Concerning the fluids and the flow, some fluids carry solid particles which may obstruct some of the pores diminishing their size or even closing them, some others may chemically and physically react with the medium enlarging the pores; so permeability changes during time and the flow occurs as if the medium had a memory. The scope of this presentation is to show, with experimental data, that the permeability of sand layers may decrease due to reassessment of the grains and consequent compaction as shown qualitatively by Elias and Hajash [1992], He [2001] and He et al. [2002]. We also provide a memory model for diffusion of fluids in porous media which fits well the flux rate observed in five laboratory experiments of diffusion of water in sand. Finally we show that the flux rate variations observed during the experiments are compatible with the compaction of sand, due to the amount of fluid which went through the grains locally, and therefore with the reduction of porosity. All the experiments have been set in the Applied Geology Laboratory at the department of Earth Sciences of the University "La Sapienza" of Rome".

#### 1. Foreword

It may seems surprising the presence in this circumstance of a relation whose title sound theoretical. I would like to say as foreword that the right word to use in order to explain this is: Theoretical Geochemistry, which has the same right of existence as Theoretical Physics and Theoretical Seismology (which I was teaching at Texas A&M University) as all the other theoretical disciplines.

A second point in favor is that the memory mechanisms, or memory formalisms, are entering in many disciplines. First it was Rheology in the Geological applications which may be developed in terms of memory formalisms. In this sector we may understand that the relaxation of the stress being accumulated tectonically is an important factor to take into account for instance in the case of the preparation of earthquakes.. In this case it is seen that using reliable data the time of accumulation of the elastic energy needed to release an earthquake is about the double that measured on the surface of the Earth without taking into account rheology.. This is one of the causes which renders difficult the forecast of earthquakes: we cannot make a reliable estimate of the time required to generate an earthquake until the rheological phenomena are taken into account.

The same may be repeated for the phenomena occurring in economy. According to Schumpeter it is not possible to guide economy with monetary instruments, for instance the prime rate. The reason being that the response times of the operators in economy when the prime rate changes.

An analogous discussion is valid also for biological phenomena.

In any case my presentation here will not be theoretical because I am an experimentalist and I will talk of the experimental aspects of the work done. I could never understand why it is sufficient to solve a differential equation system to be called "theoretician"; sometimes in a diminishing way or with the intention to diminish the value of the work done.

The entering of memory in many disciplines, according the theory of the Greek philosopher Plotino could be taken as a sign of decadence but it not so because in our case it is a base for the construction of future developments.

In fact my presentation will deal with the experiments which we have done, in the Departments of Earth Science of the Università La Sapienza od Rome, on the diffusion of water in porous media.

To introduce the mathematical memory operators we begin with the definition of the quality factor

(1)

 $dE/2\pi E = 1/Q$ 

which, in a periodic motion

$$f(t) = exp(-at)sin(\omega t)$$

gives

 $1/Q=2\pi/\omega$ 

and since

 $\varpi = 2\pi/T$ 

may be written

1/Q = aT

That is doubling the period of a pendulum we double the dissipation per unit cycle which may seem reasonable because doubling the duration of a cycle we would double the dissipation. But it is clearly not so because doubling the period, that is making the length of the pendulum longer, its velocity diminishes and with it diminishes the turbulence in the medium surrounding the pendulum and then, with the same amplitude, diminishes the dissipation per unit cycle.

Considering that the relation (1) may be representative of an exponentially decaying oscillation in an anelastic medium, whose stress is strain relation contain a first order derivatives of the strain and/or of the stress, we see that the resulting quality factor is inconsistent with experimental results. The same is true, as we already mentioned, also in the rheological models resulting from these stress strain relations. A more complex stress strain relation is needed to represent the observed quality factor. According to Kornig and Muller the rheological model which better represents the behavior of the Earth, for instance the postglacial rebound, is that which uses Fractional Order Derivatives (FOD).

### 2. Introduction

Concerning the correlation between seismogenesis and fluid migration and/or injection in the

ground, is well know the case of the Rangeley Colorado experiment (Raileigh, et al. 1976). Other important cases are the interpretation of the phases between the water level increase in reservoirs and the seismicity (Bell & Nur 1978), the stability of faults induced beneath a reservoir with cyclic variations in water level (Reoloffs 1988), the persistent water level changes in a well near Parkfield, California, due to local and distant earthquakes (Roeloffs 2001), the creep-rate changes at Parkfield, California 1966-1999: seasonal, precipitation-induced, and tectonic (Roeloffs 2001), on water level changes induced by local and distant earthquakes at Long Valley caldera, California, (Roeloffs et al. 2003), the seismically-induced water-level oscillations in a fractured- rock aquifer well near Grants Pass, Oregon (Woodcock and Roeloffs 1996), the water level changes in response to the December 20, 1994 M4.7earthquake near Parkfield, California (Quilty, and Roeloffs 1997), the hydrologic response to earthquakes in the Haibara well, central Japan, including groundwater-level changes, atmospheric pressure, rainfall, and tidal responses (Matsumoto et al. 2003), the hydrologic response to earthquakes in the Haibara well, central Japan: II, inferred from time-varying hydraulic properties (Matsumoto and Roeloffs 2003) and other numerous cases which still present unsolved problems which may find better interpretation of the data when using a more complex diffusivity model than that based strictly on Darcy's law.

In recent times, in the scientific literature, the role of diffusion of underground fluids in many fields of applied and theoretical research has been rapidly increasing. In all cases is clear that, in order to establish an accurate correlation between the observed phenomena and the migration of underground water, we should have better models of the diffusion of fluids in rocks.

The studies of the diffusion of water in porous media have therefore become increasingly numerous and varied in terms of approach; however the majority of them has devoted the attention to the diffusion of the pressure of the fluid (Barry & Sposito 1989; Kabala & Sposito 1991; Neuman & Shlomo 1993; Dewers & Ortoleva 1994; Hu & Cushman 1994, Indelman & Abramovich 1994; Steefel & Lasaga 1994; Christakos *et al.* 1995; Mainardi *et al.* 1996; Cushman & Moroni 2001; Moroni & Cushman 2001). One reason is that the law of Darcy, in most cases, allows to obtain the flux. directly from the pressure gradient, while in the case of presence of memory one is supposed to go through the somewhat complicated algebra of fractional differentiation. However in the latter case, the use of the Laplace transform domain allows, as we shall see, to obtain directly the flux when the boundary conditions on the pressure are given, which is the most common case. As in the classic case of pure Darcy's law, the equation governing the flux is the same as that governing the pressure.

The discussion of the diffusion of the fluid which we will attempt in this presentation will then be made also by transferring part of the mathematical formalisms to the frequency domain.

Many authors contributed in various forms, using Darcy's law which states that the flux is proportional to the pressure gradient, to set equations rigorously representing the interaction between the porous media and the flow of fluid through it and obtained equations solutions in many interesting cases [Bear, 1972; Sposito, 1980; Steefel and Lasaga, 1994; Dewers and Ortoleva, 1994; Indelman and Abramovici, 1994; Mainardi et al., 1998; Cushman and Moroni, 2001; Moroni and Cushman 2001]. In spite of this, some data on the flow of fluids in rocks exhibit properties which may not be interpreted with the classical theory of propagation of pressure and fluids in porous media [ Bell and Nur, 1978; Roeloffs, 1998 Fault stability) with many of the new theories.

Concerning the fluids and the flow, some fluids carry solid particles which may obstruct some of the pores diminishing their size or even closing them, some others may chemically and physically react with the medium enlarging the pores; so permeability changes during time and the flow occurs as if the medium had a memory, intending that at any instant the process of diffusion is also affected by the previous local value of pressure and flow of the fluid. This phenomenon would be taken into account when writing equations for diffusion of fluids in porous media.

The scope of this presentation is to show quantitatively, with experimental data, that the permeability of sand layers may decrease due to reassessment of the grains and consequent mechanical compaction [Elias and Hajash, 1992, He 2001, He et al. 2002].]. We will also provide, by rewriting the constitutive equation of diffusion with memory formalism, a new model for diffusion of fluids in porous media [Caputo, 2000] in order to describe permeability changes observed in the flux rate through the sand samples.

The classic theory, in the case of constant diffusivity, with constant boundary and initial conditions, would give a constant flux contrary to the results of our laboratory experiments. One would have to introduce in the equations a time variable diffusivity which is a priory unknown and would have to be determined monitoring the permeability changes caused by the flux in the sand.

#### 3. The flux of water in porous media. The experimental results.

The data are limited to about 10 hours for practical reasons. In the first few hours the flux rate steadily decreases defining a transient phase. It seems that in several hours, seemingly less than 10 hours, after the transient phase, the flux stabilises but we cannot rule out that asymptotically the flux is nil.

If the flux were constant after 10 hours then the rigorous solution requires that c = = 0 in the constitutive equations [Caputo 2000] which implies that asymptotically the flux is constant as required by Darcy's law. We have then two options

1) consider that the transient phase is asymptotically nil

2) consider that after the transient phase the flux stabilises.

However, since we have no indication of the asymptotic value and for simplicity of computation, we studied only the transient phase and use the model with c = 0.

In all experiments we have observed that flux decreases in time to about 71% of initial value and that the volume of sand reduces of about 3%; moreover, using empirical Fair and Hatch law for permeability, the sand volume and flux reductions seem compatible; which proves that mechanical compaction occurring during diffusion is cause by the permeability changes which in turn cause the flux variations.

Note that for each experiment the value of the minimum AD (the parameter minimized in the data fitting to the theoretical curve (Iaffaldano et al. 2003)) numerically computed is about 2% of the average observed flux and that the order of the fractional derivatives has a standard deviation of 0,048 (or 9%) of the average value obtained in the different experiments which, taking into account the variety of samples, is rather satisfactory and, with the low value of AD, confirms the validity of the model.

We have also seen that, with the boundary and initial conditions used, the relaxation time of the flux, that is the time to reach stability, is about 10 hours which in turn implies that the compaction of the sand in the sample has the same relaxation time.

However in terms of the memory model the flux and the associated relaxation time are now defined by two parameters, and not only one as in the classic theory; the parameters are the order of fractional derivative *n* and  $d\mu/\gamma\rho_F$ , where  $\mu$  is the viscosity of the fluid, which are called pseudo-diffusivity [Caputo, 2000].

The constitutive equations of the flux is where the fractional derivative of order n is

$$\gamma \overline{q}(\overline{x},t) = -\left[c + d\frac{\partial^n}{\partial t^n}\right] \overline{\nabla} p(\overline{x},t)$$
$$ap(\overline{x},t) = \alpha \rho(\overline{x},t)$$

where the fractional derivative of order n is

$$f^{(n)}(t) = \partial^n f(t) / \partial t^n = \frac{1}{\Gamma(1-n)} \int_0^t \frac{\dot{f}(u)}{(t-u)^n} du$$

where n is a real number  $0 \le n \le 1$ . The solution at the extreme x = l of the layer is

$$q(0,t) = -\frac{dBK}{2\pi\gamma} \int_{0}^{+\infty} \frac{e^{-rt}}{r^{\nu}} \cdot \frac{2\sin(\pi\nu) e^{2Mr^{\nu}} - 1 + 4\sin(Nr^{\nu})\cos(\pi\nu) e^{Mr^{\nu}}}{e^{2Mr^{\nu}} + 1 - 2\cos(Nr^{\nu}) e^{Mr^{\nu}}} dr$$

K =constant pressure difference across the sand layer

$$M = 2Bl\cos(\pi\nu); \qquad N = 2Bl\sin(\pi\nu)$$

 $B = [\gamma \alpha / \alpha d]^{1/2}$ v = (1-n)/2. $a / \alpha = \rho_F z / k_F$ 

Values of the memory parameters  $(n, d/\gamma)$  of the curves fitting the data in the five experiments.

	n	$\frac{d}{\gamma}(s^{1+n})$	$AD(g \cdot s^{-1})$	$q_{\scriptscriptstyle AS}($
Exp. 1	0.46±0.01	$0.008 \pm 0.001$	0.8	30.3
Exp. 2	0.58±0.01	0.014 ± 0.002	0.41	27.1
Exp. 3	0.54±0.01	0.012 ± 0.002	0.52	27.5
Exp. 4	0.54±0.01	0.010±0.001	0.55	27.2
Exp. 5	0.58±0.02	0.046±0.003	0.8	27.1

 $q_{AS}$  = value of the flux at the end of the experiment.

In order to best fit memory model to experimental data we minimized the following two variables function

$$AD\left(\nu,\frac{d}{\gamma}\right) = \frac{1}{N_D} \sum_{i=1}^{N_D} \left| ED_i - q\left(t_i,\nu,\frac{d}{\gamma}\right) - q_{AS} \right|$$
(19)

where  $N_D$  is the number of experimental data for each experiment,  $ED_i$  are the data obtained in the laboratory at the time  $t_i$ .

Glossary  $g \cdot cm^{-3}$ Mass of sand per unit volume  $\rho_s$  $cm^2$ k Permeability z [dimension Porosity  $\overline{[ess]}$  $q(x,t) \quad [g \cdot s^{-1} \cdot cm^{-2}]$ Fluid mass flow rate in porous medium  $p(x,t) \quad \left[g \cdot s^{-2} \cdot cm^{-1}\right]$ Pressure of the fluid  $\rho(x,t) \quad \left[g \cdot cm^{-3}\right]$ Variation of fluid mass per unit volume in porous medium from the undisturbed condition  $\rho_F \qquad \left[g \cdot cm^{-3}\right]$ Mass of fluid per unit volume  $k_B \qquad g \cdot s^{-2} \cdot cm^{-1}$ Bulk modulus of fluid

 $\begin{array}{c} \mu \\ g \cdot s^{-1} \cdot cm^{-1} \end{array} & \text{Viscosity of fluid} \\ \\ \hline \frac{d\mu}{\gamma \rho_F} & \left[ s^n \cdot cm^2 \right] & \text{Pseudodiffusivity} \end{array}$ 

## 4. The case of periodic flux inversion.

An interesting case arises when the direction of the flux is periodically inverted as, for instance, when it is caused by the ocean tides (Robinson 1939). The shortest period of the lunar and solar tides are around 12 and 24 hours which are somewhat shorter than the duration of our experiments which however show that the settling of the sand grains takes less than 10 hours. It is then clear that the flux caused by the migration of waters underground settles the grains of sand before the next cycle begin and then the average flux rate is close to the average value of the flux rate observed in the laboratory in the 12 hours components and close to the asymptotic value observed in the 24 hours tidal component; obviously the values of the observed flux rate must be normalised to the conditions and the amplitude in the field.

### **Figure caption**

Figures a) and b). Grain size distribution of sand samples.

Figure 2. The experimental device

Figures 4.6, 4.7 and 4.8: The flux across the sand samples is represented by the dots. The solid curve is the fitting of the data produced in one of the five experiments to the theoretical curve produced with the memory model of the flux.

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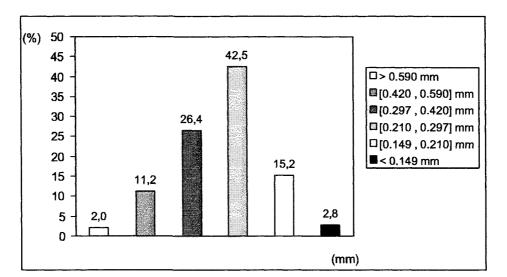
Roeloffs, E., M. Sneed, D.L. Galloway, M.L. Sorey, C.D. Farrar, J.F. Howle, J.Hughes, *Water level changes induced by local and distant earthquakes at Long Valley Caldera, California*, in press, J. Volc. Geotherm. Res., 2003.

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Sand density was estimated to be  $\rho_s = (2.4 \pm 0.1) \frac{g}{cm^3}$ .

We used water as fluid, its temperature during all experiments was  $(19 \pm 1)$  °C.





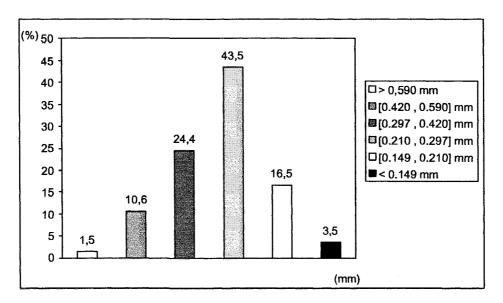


Figure 1.b

A schematic description of the instrument assembled for the diffusion experiments is shown in Figure 2.

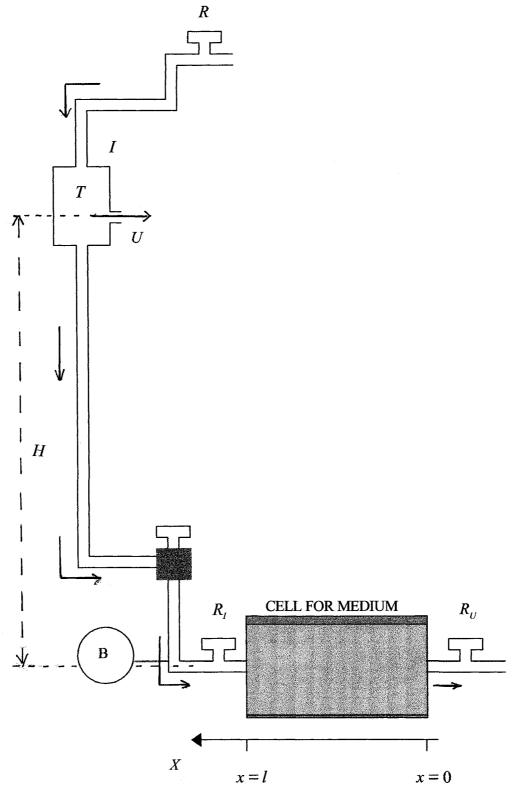


Figure 2 : experimental device

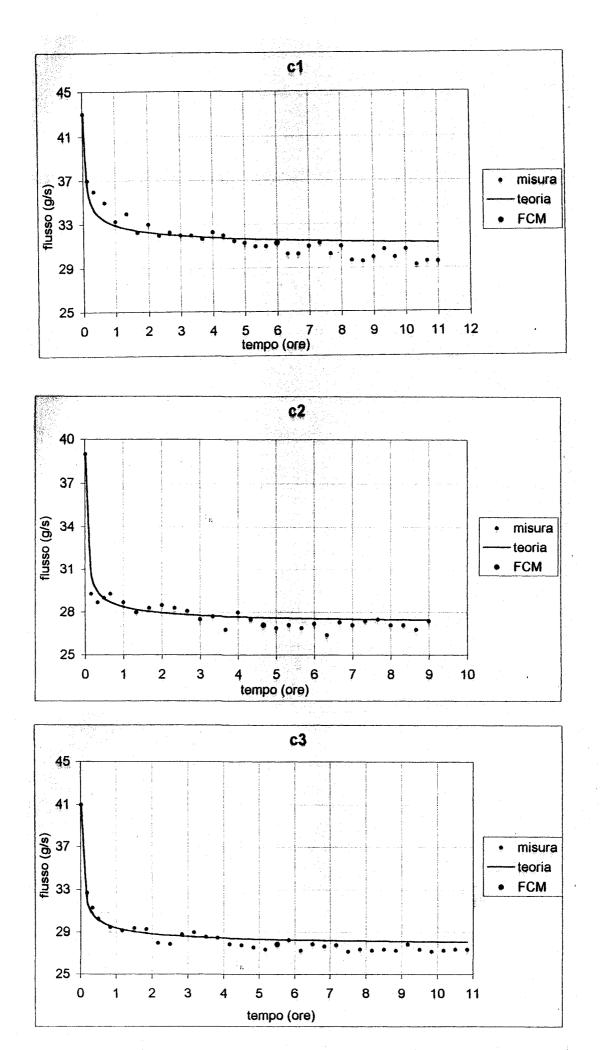
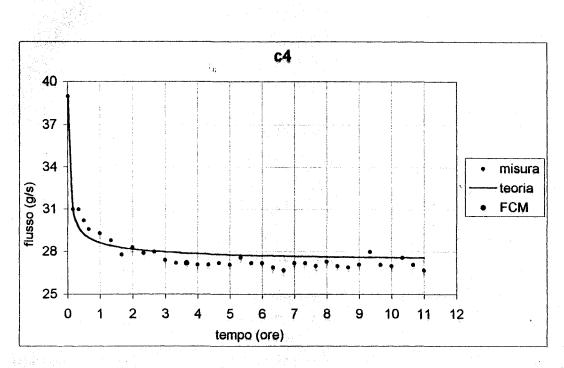


Fig. 4.6





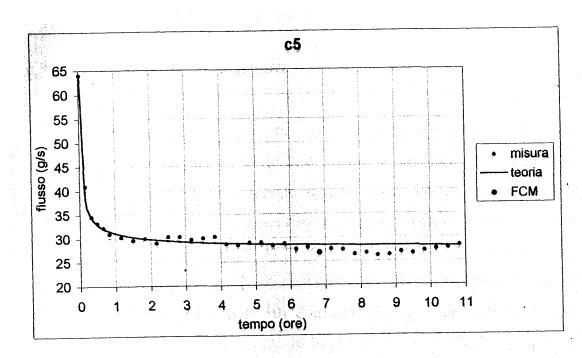


Fig. 4.8