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RESEARCH ARTICLE

Entrapment of hydrocarbon oil in water-saturated sediment: implications on hydraulic properties

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Abstract

The infiltration and the entrapment of hydrocarbon oil on a layer of sediments made up of sand are analyzed in experiments by a device of Darcy. It is shown that the permeability and the porosity of the porous environment are modified. It also discusses the changes in the viscosity of the pollutant after passing on the sand. The analysis of the whole of the experimental results makes it possible to analyze the trapping of the pollutant in the sedimentary layers according to their granulometry. The pollutant which we believe has in its physico-chemical composition of long chains. The impact of the infiltration of the pollutant on the modification of these components will be discussed.

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INTRODUCTION

Due to maritime traffic, the nearshore is exposed to the risks of pollution by hydrocarbons. Through infiltration or mixing processes, such products may be present in the sedimentary layer for long periods and then be moved and transported in the flow by hydrodynamic forcing. During big storm events, this transfer to the fluid is mainly due to the sandy bed erosion, at the origin of oil migration towards the fluid. During fair weather conditions, or in sheltered zones, sewage of the saturated water table during tidal cycles may drain hydrocarbons to water.

Such kind of study has drawn interest for its practical interest in underground-water pollution by organic solvents or oil products. Either microscopic or macroscopic approaches have been proposed for the study of the two phase flow within unconsolidated porous media or consolidated sandstones. (Gioia and Urciuolo, 2006; Gioia et al., 2003), investigated experimentally the mobilization of an oil bank under the combined effect of Bond (NB) and capillary (NC) numbers, in a packed bed column of glass beads saturated with water. For still water (Gioia et al., 2003), they found migration conditions according to NB values. In the presence of the flow of water, they found the oil mobility to depend on the vectorial sum of NB and NC. In order to reach the irreducible saturation, the experiments have been run with sweeping water velocities outside the range of validity of the Darcy's law.

Experiments on the relative permeability of water (k_{rw}) and oil (k_{ro}), as a function of the capillarity number Ca have been carried out in both steady-state (Tsakiroglou et al., 2007) and transient flow conditions on a same porous medium (Rangel-German et al., 2006). It was found that both the wetting and the non-wetting phase relative permeability depend on both the flow regime (transient or steady state) and on the wettability of the two-phase flow. Either transient growth pattern or steady-state flow regimes may explain differences observed for the steady-state and for the transient relative permeabilities. For fractured systems, Rangel-German and al. (Rangel-German et al.,

2006) found that permeability depends on both capillarity and viscous forces. In the case of oil-water drainage, oil saturation is the higher near the small fractures, and fingering is observed in the wide fracture.

In the nearshore, oil spill from maritime traffic may settle within the sediment. (Idrissi, 2006) proposed a synthesis on the various types of hydrocarbon pollutants suitable to be trapped in the sand. For a better understanding of oil trapping and moving within the sediment layer, oil infiltration and trapping within sandy beds of various granulometry is experimentally studied through a macroscopic approach. Sediment water permeability is first investigated in saturated conditions for porous media made of successive sandy layers of different grain size. Oil fixing and drainage are then investigated first through biphasic oil-water flows, and second after water drainage. Analysis and conclusions are based on the characteristic parameters (permeability, porosity, viscosity and organic matter) measured during the laboratory experiments.

Materials and methods:

Experimental material:

A sketch of the experimental device is presented in figure 1. The porous media is located in the stainless steel vertical cylinder, of inner diameter $D=0.073\text{m}$, of total length $L=0.350\text{m}$. The liquids are injected at the top of the cylinder. A manometer allows pressure gradient measurement. In the experiments, the maximum flow rate was $Q_{\max}=6.8/\text{min}$ (velocity $V_{\max}=0.028\text{m/s}$), and the maximum pressure difference between the top and the bottom of the porous media was $\Delta P=550\text{ Pa}$.

In the biphasic cases, 10ml of oil was mixed with 100ml of water just above the porous media entrance. The SAE (Society of Automotive Engineers) classification of the oil was 40W20; its viscosity in the experimental conditions was 4650 cP (Bouamrani et al., 2008).

The porous media:

The sediment was collected on the Mohammedia beach (Morocco), and then sifted and sorted. The porous media is made of a layer or successive layers of equal length of sand of given class (Idrissi, 2006). The average diameter is given in table 1.

The studied samples consist of one, or two or three successive layers of given classes of permeability K (Table 2). For a porous media of n layers of equal thickness $L_n=L/n$, $n=2,3$, the equivalent permeability is defined respectively by:

$$(1) \quad k_{e2} = \frac{2k_1k_2}{k_1 + k_2}, \quad k_{e3} = \frac{3k_1k_2k_3}{k_1k_2 + k_1k_3 + k_2k_3}$$

With K_e : Equivalent permeability, K_i the permeability of layer i , $i=1,\dots,n$.

Results:

Three successive stages are considered for the experiments. In a first stage, monophasic flows are considered for the calculation of water permeability of the porous samples. Oil fixing is then investigated through biphasic oil-water flow across the porous. Drainage of the porous media with water is then considered in a last stage. Macroscopic parameters and their evolutions are presented in the following.

Permeability:

The permeability of the sedimentary layer is first determined for water flow. Various flow rates up to 6.85 l/min showed a linear behavior of the flow rate Q with respect to the pressure difference ΔP .

The Darcy law:

$$Q = \frac{Sk}{\mu} \frac{\Delta P}{L} \quad (2)$$

S is the section of the cylinder, $\Delta=10^{-3}\text{ Kg.m}^{-1}.\text{s}^{-1}$ the water dynamic viscosity, was then applied for the calculation of the permeability k .

The water viscosity was also used for the calculation of the permeability for oil-water flows, even if viscosity is slightly affected by the presence of oil. However, this choice was motivated by the non-miscibility of the two

phases, and their different behavior within the sediment according to the size of the pores. The permeability as a function of the porous media is presented in figure 2 for the monophasic flow, the diphasic flow and the drainage. Theoretical equivalent permeability for multi-layers media is also presented for the monophasic flow.

We can observe that the permeability depends slightly on the order of the layers, but near the equivalent value given in table 1.

The permeability is strongly affected by the presence of oil in the water, with a decrease of the permeability by a factor of 10. Quasi-reversible behavior is observed for the coarse grains, for which initial permeability is almost recovered after drainage. For porous media containing medium or fine grains, the permeability increase again after drainage but remains much closer its value before contamination.

Porosity and time of residence:

A viscous meter Brookfield was used to determine porosity. Porosity of the sediment before and after oil contamination is presented in figure 3 for the four sediment classes. We can observe a regular decrease of the porosity from 42% to 27% from coarser grains to finer grains. After contamination, porosity is lower, especially for the finer grains with a decrease by a factor of 2, due to oil trapping in the sediment.

The time of transit of oil across the porous media is presented in figure 4, before and after oil contamination. We can observe that in the latter case, duration is increased nearly 2 to 3 times. Let us note that a strong increase is also observed for the coarser grains, even if porosity is only slightly inferior (Figure 3).

Oil viscosity and organic matter:

The contaminant used is hydrocarbon oil of viscosity 4650 cP (Idrissi, 2006). After migration across the porous media, viscosity has decreased as shown in figure 5. The maximum decrease is observed for the finer sands. We can also notice that the order of the sediment layers has poor influence on the viscosity after oil transit. We can also observe that the decrease is higher for successive layers of coarse and very coarse sand layers than for either coarse or very coarse sand alone.

In order to have a quantitative idea of the oil trapped, the Walkley Black oxidation method was used to determine organic matter within the porous media (Idrissi, 2006). As shown in figure 6, a huge increase of the organic matter within the sediment is observed after contamination. A good correlation is thus observed with the viscosity decay (Figure 5)

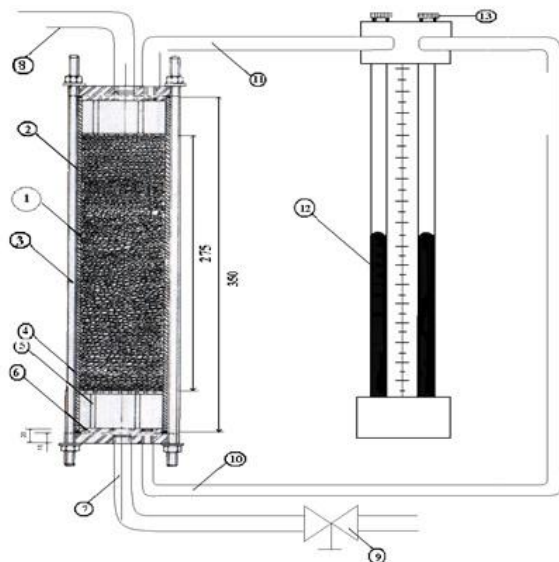


Fig. 1: Simplified diagram of the experimental device: 1- Cylindrical body, 2- Porous environment, 3- Threaded rod, 4- Strainer, 5- Stem support strainer, 6- Base plate, 7- Inlet pipe, 8- Out let pipe, 9- Winnow, 10- Entered pressure tap, 11- Left pressure tap, 12- Pressure gauge with mercury, 13- Bleed hole.

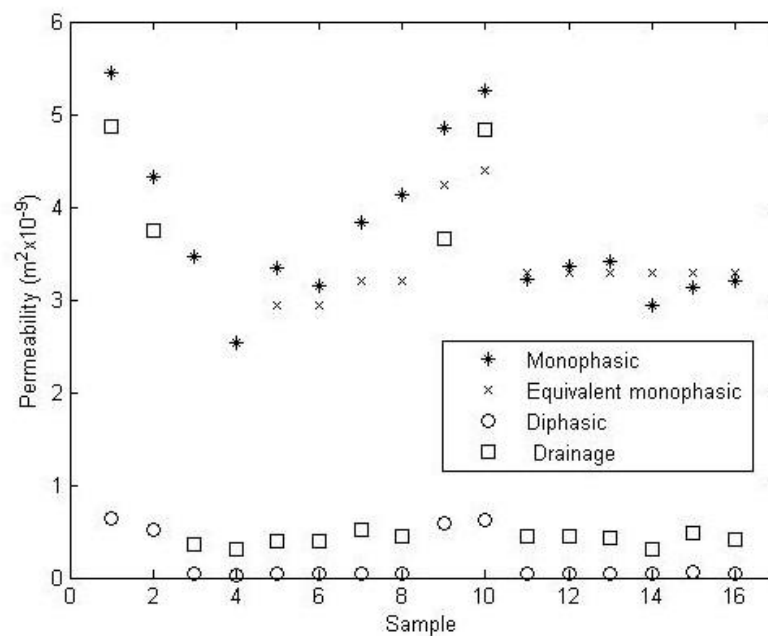


Fig. 2: Comparison of the permeability for the flow monophasic, diphasic and monophasic after the rinsing.

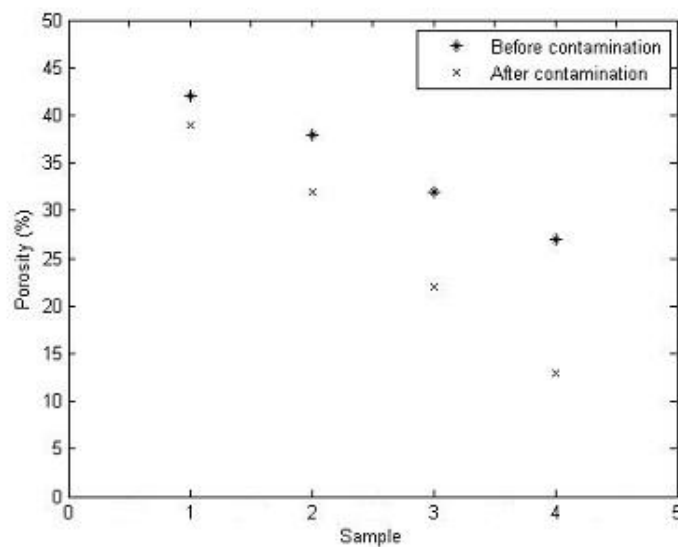


Fig. 3: Measurement of the porosity before and after contamination for the four sand samples.

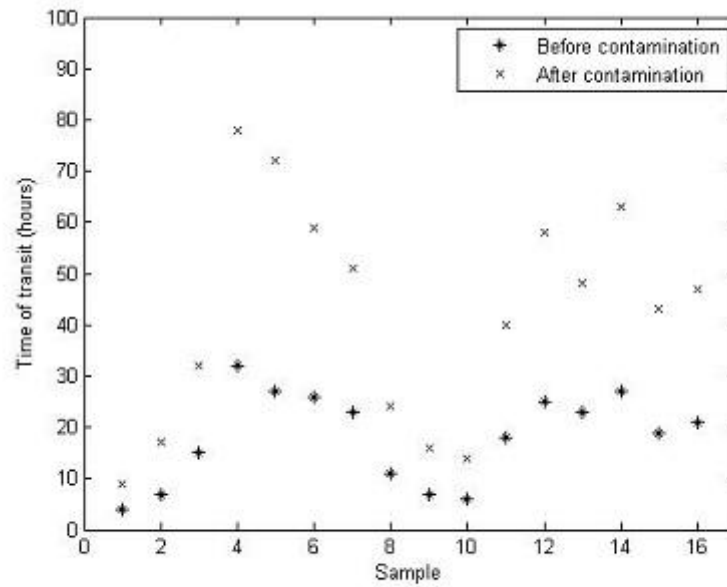


Fig. 4: Measurement of the trapped volume and the time of transit in function of the diffusion before and after contamination for the four sand samples.

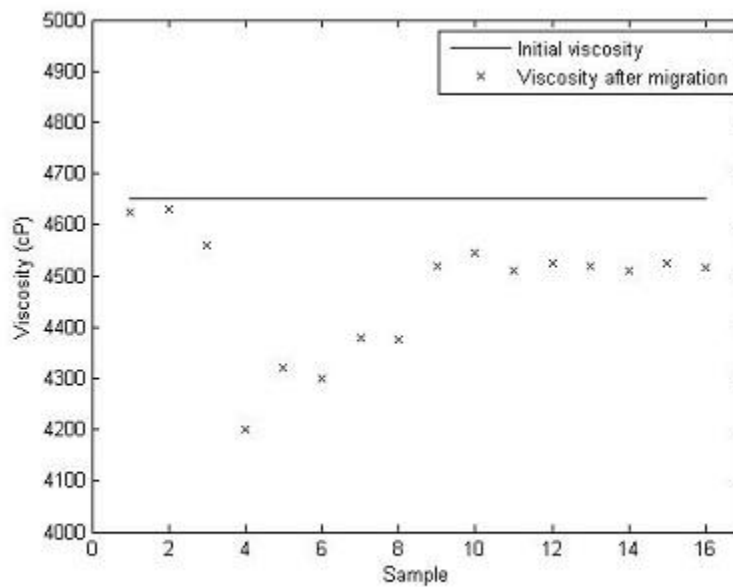


Fig. 5: diagram of viscosity evolution of the pollutant before and after the transit for the four sand samples.

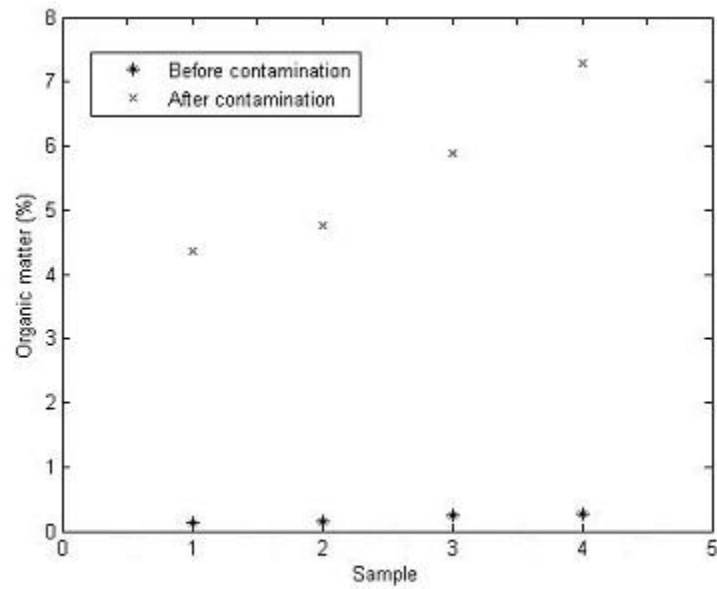


Fig. 6: Matter organic evolution in function of the porosity for the four sand samples.

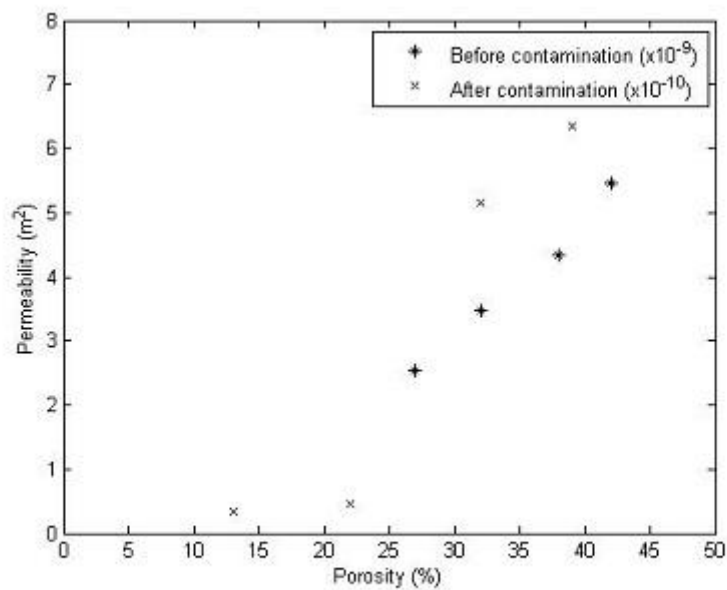


Fig. 7: Permeability Evolution in function of the porosity for the four sand samples.

Table 1: Particle-size ranges

D in mm	Particle-size ranges
1.25<D< 2.00	Very Coarse Sand (V.C.S)
0.63<D< 1.25	Coarse Sand (C.S)
0.25<D< 0.63	Medium Sand (M.S)
0.06<D< 0.25	Fine Sand (F.S)

D: Median Diameter ; mm: millimeter

Table 2: Porous media configurations

Sample	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16
Layers	VCS	CS	MS	FS	FS + MS	MS + FS	FS + CS	CS + FS	VCS + MS	MS + VCS	FS + MS + CS	FS + CS + MS	MS + FS + CS	MS + CS + FS	CS + MS + FS	CS + FS + MS

CS: Coarse Sand; MS: Medium Sand; FS: Fine Sand; VCS: Very Coarse Sand

Discussion:

As shown in figures 2 and 3, both permeability and porosity decrease with sediment size, independently of oil contamination. However, as shown in figure 7, the same proportionality between permeability and porosity is observed whatever the sediment size for the monophasic case, contrary to the oil-water case where different trends are observed either for finer or coarser grains. Indeed, for coarser grains, similar relationship is observed between permeability and porosity, which means that oil particles are adsorbed at the grain surface, without pore obstruction. This hypothesis is confirmed by the permeability calculation after drainage, which shows that initial permeability is recovered after drainage for coarse and very coarse grains samples (Figure 2).

On the contrary, oil contamination seems irreversible for the finer grains. Relationship between permeability and porosity for oil-water case seems to correspond to porous obstruction. This hypothesis is confirmed by the weak increase of the permeability after drainage, far from the initial value calculated in the monophasic case (Figure 2). The increase of the time of residence within the porous media after contamination shows that the oil chains play an important role in the oil trapping within the sediment. The decrease of oil viscosity after migration across the porous media then corresponds to the trapping of the larger chains.

Conclusion:

The aim of the present experimental study was a better understanding of the oil infiltration, fixing and drainage within sediment porous layers. We have observed two different behaviors according to the grain size, concerning the longer chains constituting the oil. For coarser grains, the oil is adsorbed preferentially at the grain surface, and then may be easily moved through water drainage. For finer grains, the oil is fixed within the pores, and hardly moved by

water drainage. For nearshore processes, we can conclude that trapped oil may be moved under water drainage, as observed in the intertidal zone during tidal cycles. For finer sand beaches, the time of residence will be strongly increased, and oil mobilization will be significant if the sandy bottom which moved to suspension caused by the hydrodynamic forcing.

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