



DARCY SCALE NAPL DISSOLUTION IN HETEROGENEOUS STRATIFIED POROUS MEDIA

B. Mabilia¹, C. Tathy² and D. Nganga³

¹Higher National Polytechnic School, Marien Ngouabi University, Brazzaville, Congo

²Higher Teachers' Training School, Marien Ngouabi University, Brazzaville, Congo

³Department of Environment Science, Faculty of Sciences, Marien Ngouabi University, Brazzaville, Congo

E-Mail: bernamab@yahoo.com

ABSTRACT

The aim of this study is to develop NAPL dissolution in heterogeneous porous media by one and two dimensional physical model at macroscopic Darcy scale. The model describes the movement of a water phase in the presence of a trapped polluting phase at residual saturation. Two limit cases of dissolution were studied: local equilibrium case and the local non equilibrium case. The construction uses the finite volume methods for the approximation of equations. The numerical tests emphasize particularly the evolution of the zone of exchange between the organic and water phases. Local non equilibrium situations obtained following the mechanisms hereafter, heterogeneity effects of the medium and heterogeneity effects of the initial saturations induced by hydrodynamic instabilities or particular conditions of installation, were characterized.

Keywords: modelling, NAPL dissolution, heterogeneous porous media, Darcy scale, finite volume methods, equilibrium.

1. INTRODUCTION

These last years, research on the groundwater in many countries, related to the control of water quality. Since several decades, human contribute to increase the chemicals rejections in the environment. The subsoil waters are thus gradually contaminated by the human activities. To control this contamination, several restrictive laws were installed. They allowed in some countries a notable improvement of the quality of water. However, in certain cases the deterioration of the water quality is difficult to envisage. That is in particular with the presence of heterogeneities in the geological medium which disturb the fields of concentration of the pollutant, thus making the analysis very difficult. These heterogeneities of structure are met from μm -scale to the km scale. Their hierarchical organization controls the flows, the migration of the pollutants or hydrocarbons and, consequently, the transfers fluid-fluid and fluid-rock such as dissolution and adsorption. Currently, the major problem to which the scientists face in the protection of the water resources is to identify the mechanisms by which the pollutants can infiltrate in the aquifer. They seek to develop good systems of prediction of the transport of contaminant in groundwater. However, the various models which were developed produce a large variety of predictions. The cause of this variability of the results is, amongst other things the difficulties of characterization of the medium, heterogeneities of the porous media and those coming from the differential distribution of the pollutant within this media. These last years, several work was undertaken to try to explain the influence of heterogeneities on the transport of pollutants in porous media. These studies concern to a large extent, the dissolution of trapped NAPL present in the porous media. Indeed, Illangasekare *et al.*, (1995) affirm that heterogeneities of the porous media can modify the trajectory of the pollutants significantly and thus lead to a flow which can generate a local non

equilibrium situation. This assertion is supported by Mayer *et al.*, (1996) who estimate that the dissolution modeling depends on the knowledge of the heterogeneous distribution of the pollutant in the medium considered. Numerical studies (Park E and Parker J.C., 2005) on the evaluation of the kinetics of DNAPL dissolution in heterogeneous porous media made it possible to calculate the transfer coefficient in order to describe NAPL mass transfer in the source zone.

Other numerical work of dissolution modeling (Saenton *et al.*, 2002; Soga *et al.*, 2004; Côme *et al.*, 2005; Mayer A. and Endres K. L., 2007) was carried out. These numerical studies show among other things that: (i) the effectiveness of the NAPL dissolution depends on the heterogeneity of the aquifer which controls the trapping of NAPL, on the aqueous phase flow deviation resulting from heterogeneity and on the reduction of relative permeability due to the trapping of NAPL (Saenton *et al.*, 2002); (ii) the morphology of the NAPL source zone, determined by the heterogeneity of the porous media controls dissolution, in other words, the precise characterization of the site of the trapped clusters is essential to envisage dissolution flows (Soga *et al.*, 2004); (iii) when initial residual polluting saturation is discontinuous, pure water entering in the source zone comes out again with a certain concentration different or no of equilibrium concentration. Even if in the saturated zones in pollutant dissolution is of the local equilibrium type, the concentration at the exit can be significantly weaker than the equilibrium concentration. Local non equilibrium situations can thus appear following various mechanisms: heterogeneity effects at the pore-scale and heterogeneity effects of the medium or initial saturations (Mabilia *et al.*, 2003, 2004; Côme *et al.*, 2005). One can advance the following assumption: the differential dissolution of the various mediums met can still lead to zones which have low polluting phase content beside zones which have high content. Even if, at small



scale, the mechanisms of dissolution can be described by a model of local equilibrium type, it is not necessarily the case at large scale. At this scale, polluted water at equilibrium mixes with no contaminated water, and the result gives a concentration weaker than the equilibrium concentration at the exit of the source zone, situation characteristic of local non equilibrium. It emerges from this bibliographical review that heterogeneities as well of the porous media as those of residual saturations in this media, significantly take part in the dissolution of a polluting phase. It is then advisable to take them into account at the scales of modeling. It is the object of this study.

2. THEORY

Let us consider a water phase (w), polluted by a partially miscible phase (o). At the interface, at microscopic scale, the thermodynamic equilibrium condition imposes that the polluting concentration in pollutant is equal to equilibrium concentration, that we note C_{eq} . Let us define the macroscopic concentration C of α -component dissolves in water, by:

$$C = \rho_w C_{w\alpha} \quad (1)$$

Let c , microscopic concentration. The microscopic condition to satisfy by c at the interface $o-w$, (noted A_{wo}) is written:

$$c = C_{eq} \quad \text{at} \quad A_{wo} \quad (2)$$

If the gradients of concentration within the representative elementary volume (REV) are small, the microscopic concentration c is very close to equilibrium

concentration C_{eq} within the pores, and it is appropriate to write at macroscopic scale the following approximation:

$$C = C_{eq} \quad (3)$$

This approximation is called local equilibrium approximation, characterized by the fact that the microscopic equilibrium condition at the interfaces is transposed at macroscopic scale in any point of the porous medium. Thus, knowing the concentration (equal to C_{eq}), one can then use the mass balance equation to calculate the exchange term. If this should not be the case, i.e., if

$$C \leq C_{eq} \quad (4)$$

This situation is known as local non equilibrium approximation. It is necessary to determine a correlation describing the complex dynamics of the exchanges. This correlation makes it possible to solve the transport equation and to calculate the macroscopic concentration field. It will be noted that a local non equilibrium model includes the local equilibrium one as limit case, the reverse not being true. Figures 1 and 2 illustrate the fields of concentration and saturation in one dimensional case, that one can obtain, on the basis of local equilibrium or local non equilibrium model. These Figures represent: (i) initial concentration and saturation fields (broken lines), (ii) the same fields observed a certain time after sweeping by water (solid lines). The local equilibrium case is characterized by a stiff dissolution front, while the local non equilibrium case by a spread out dissolution front. Figure-3 illustrate the evolution of concentration with the downstream of the source zone as function of time the one obtain, on the basis of local equilibrium or local non equilibrium model.

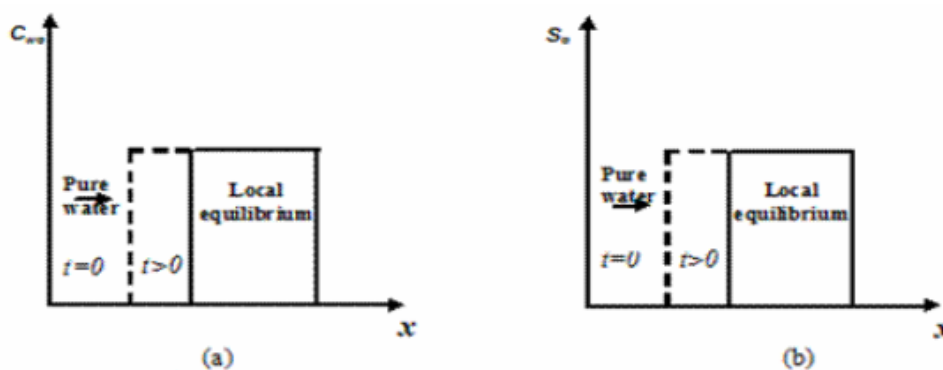


Figure-1. Evolution of concentration fields (in mass fraction) (a) and of saturation (b) in polluted phase for 1D-flow in the source zone (local equilibrium case).

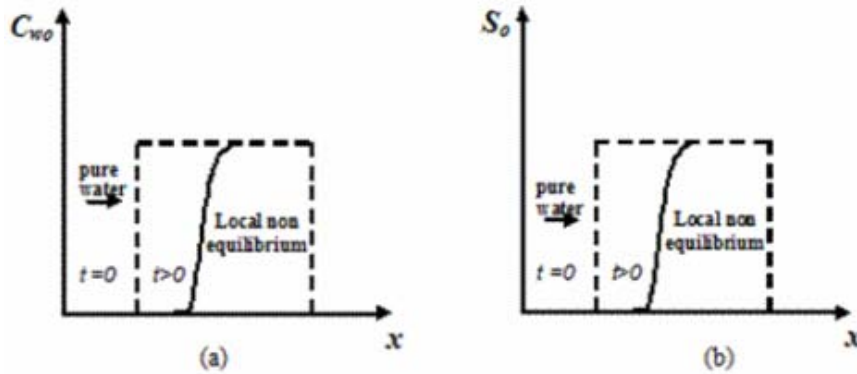


Figure-2. Evolution of concentration fields (in mass fraction) (a) and of saturation (b) in polluted phase for 1D-flow in the source zone (local non equilibrium case).

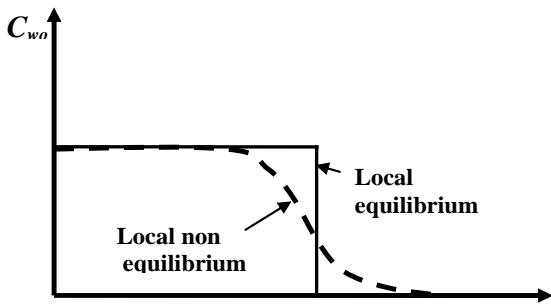


Figure-3. Evolution of concentration fields (in mass fraction) with the downstream of source zone as function of time.

3. DARCY SCALE DISSOLUTION MODELING

3.1 One dimensional physical model

The situation to be modeled is illustrated on Figure-4. We want to simulate starting from a simple modeling, the behavior of a heterogeneous porous medium, considered as representative of a type of heterogeneities frequently met the stratified model. In such a model, the medium is compared to a succession of parallel layers with constant thickness, and various

characteristics (porosity and permeability in particular). The porous medium considered in this study is in deformable and contains initially two incompressible liquid phases' oil and water. We define by unit cell a group of two different strata (ω , η), of length l_ω and l_η . The medium contains partially miscible pollutant initially, at residual saturation S_{or} . Pure water is injected perpendicularly to the strata, upstream from $x = 0$, with constant velocity V_w . In the local equilibrium case, the dissolution front is located by x_f . Each stratum is with constant porosity ϵ_ω and ϵ_η , respectively.

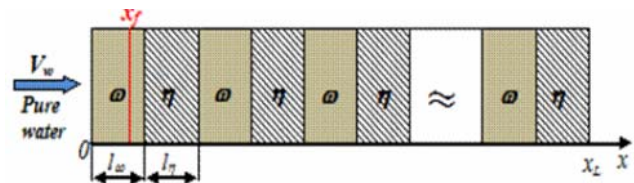


Figure-4. Physical presentation of 1D-problem.

The one dimensional equations (Quintard M. and Whitaker S., 1994) are:

$$\epsilon \frac{\partial((1-S_o)C_{wo})}{\partial t} + V_w \frac{\partial C_{wo}}{\partial x} = \frac{\partial}{\partial x} \left(\epsilon(1-S_o)D_w \frac{\partial C_{wo}}{\partial x} \right) - \alpha(C_{wo} - C_{woeq}) \tag{5}$$

$$\epsilon \frac{\partial S_o}{\partial t} = \frac{\rho_w}{\rho_o} \alpha(C_{wo} - C_{woeq}) \tag{6}$$

$$C_{wo}(x,0) = C_{woeq} \quad ; \quad S_o(x,0) = S_{or} \tag{7}$$

$$C_{wo}(0,t) = 0 \quad ; \quad \frac{\partial C_{wo}}{\partial x}(x_L,t) = 0 \tag{8}$$

where C_{woeq} , S_{or} and x_L are respectively the NAPL equilibrium concentration in the liquid phase, NAPL

residual saturation and the characteristic length of the studied field. Moreover, the mass fractions and saturations satisfy the following constraints:

$$C_{ww} + C_{wo} = 1 \quad ; \quad C_{ow} + C_{oo} = 1 \quad ; \quad S_w + S_o = 1 \tag{9}$$

3.2 Two-dimensional physical model

The situation to be modeled is the same one as that presented previously with the difference that here the strata are laid out horizontally and water is injected parallel to the strata as illustrated in Figure-5. The model is thus two-dimensional and the physical conditions of stratified medium are the same ones (constant thickness,



and various characteristics, different porosity and permeability in each stratum, indeformable porous medium containing initially two incompressible liquid phases oil and water). The unit cell includes two different strata (ω, η), of length x_L . Each stratum contains initially partial miscible pollutant, at residual saturation S_{or} . Pure water is injected upstream from $(0, y)$ axis and with Darcy velocity. It loads with pollutant by dissolving the trapped phase, and reaches a certain concentration. The studied system is a binary system for which we adopt the same conditions at the interfaces between strata and the same notations as into one dimensional case: two phases, water w and oil o containing each one two components water (w) and NAPL (o). Each stratum has constant porosity and permeabilities, respectively.

$$\frac{\partial(\varepsilon(1-S_o)C_{wo})}{\partial t} + \nabla \cdot (\mathbf{V}_w C_{wo}) = \nabla \cdot (\varepsilon(1-S_o)\mathbf{D}_w \cdot \nabla C_{wo}) - \alpha(C_{wo} - C_{woeq}) \quad (10)$$

$$\varepsilon \frac{\partial S_o}{\partial t} = \frac{\rho_w}{\rho_o} \alpha (C_{wo} - C_{woeq}) \quad (11)$$

$$\mathbf{V}_w = - \frac{k_{rw}(S_o)}{\mu_w} \mathbf{K} \cdot \nabla P_w \quad (12)$$

$$\nabla \cdot \left(- \frac{k_{rw}(S_o)}{\mu_w} \mathbf{K} \cdot \nabla P_w \right) = 0 \quad (13)$$

In the equations above, the tensors of dispersion \mathbf{D}_w and of permeability \mathbf{K} have the form:

$$\mathbf{D}_w = \begin{bmatrix} D_{xx} & D_{xy} \\ D_{yx} & D_{yy} \end{bmatrix} \mathbf{K} = \begin{bmatrix} K_{xx} & 0 \\ 0 & K_{yy} \end{bmatrix} \quad (14)$$

and Eq. (12) is the Darcy law. The dispersion tensor components are defined by (Bear, 1961, 1972):

$$D_{ij} = (\alpha_T \|\mathbf{V}_w\| + D^{eff}) \delta_{ij} + (\alpha_T - \alpha_L) \frac{V_{wi} V_{wj}}{\|\mathbf{V}_w\|}$$

where α_L and α_T are the longitudinal and transverse local dispersivities, V_{wi} the i -th component of local filtration velocity vector and D^{eff} is the local-scale effective diffusivity.

The permeability tensor components are given by: $K_{ii} = k_{rw}(S_o)k_{ii}$ $i = 1, 2$, where k_{ii} are the components of intrinsic permeability tensor of the porous medium and $k_{rw}(S_o)$ is the water phase relative permeability.

The boundary conditions (Figure-5) are:

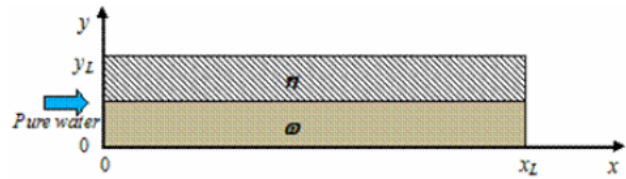


Figure-5. Physical model of 2D-problem.

The flow is two-dimensional and horizontal, the movements of the particles due to gravity are supposed to be negligible compared with those due to the pressure. The equations of this model are:

a) at north and south boundaries of the domain: we impose a condition of free flux on pressure and concentration:

$$\mathbf{n} \cdot (\mathbf{V}_w C_{wo} - \varepsilon(1-S_o)\mathbf{D}_w \cdot \nabla C_{wo}) = 0 \quad (15)$$

$$\mathbf{n} \cdot \left(- \frac{k_{rw}(S_o)}{\mu_w} \mathbf{K} \cdot \nabla P_w \right) = 0 \quad (16)$$

b) at east boundary of domain: a Dirichlet condition is used for the pressure and that of Danckwerts (1953) for diffusive flow:

$$\mathbf{n} \cdot (\varepsilon(1-S_o)\mathbf{D}_w \cdot \nabla C_{wo}) = 0 \quad (17)$$

c) at the western boundary of the domain: the Dirichlet condition as well for the pressure as for the concentration.

To discretize the term of dispersion, one used the 9 points finite volume scheme developed by Edwards and Rogers (1994), for the taking into account of the full permeability tensors in the resolution of Darcy equations, and by Cherblanc *et al.*, (2003) for the description of dispersion in a two-medium model in heterogeneous media. A 9 points finite volume scheme allows us to take into account the effect of the outside diagonal terms of the tensor, increases the precision and reduces the grid orientations effects (Urgelli, 1998).

4. NUMERICAL EXPERIMENTS

The NAPL dissolution equations in porous media were solved numerically and we present the results of the various experiments carried out in 1D and 2D. The porous media considered in this study are fine sandstones and sands.

4.1 One-dimensional experiments (1D)

The dissolution modeling in the domain illustrated on Figure-4 contaminated by a NAPL (here



TCE) was carried out using the program DISSOL 1D that we built. Two cases were studied: (i) the first case relates to the application of the assumption of local equilibrium at Darcy scale, and (ii) the second relates to the assumption of local non equilibrium.

The first case corresponds to dissolution in water of a pollutant (TCE) in a stratified porous medium of

length $XL = 20$ cm (or $XL = 10$ adimensional) containing several unit cells as illustrated in Figure-4. This dissolution is controlled by a large Damköhler number (rather large mass transfer coefficient) so that the dissolution front is stiff and representative of the local equilibrium model. The numerical simulations were carried out using the data consigned in Table-1.

Table-1. Data for the experiments relating to local equilibrium assumption.

Stratum	Porosity (ϵ)	Residual saturation in TCE (S_{or})	Mass transfer coefficient α (s^{-1})	Physical properties of the phases
ω	0.38	0.25	4	$\rho_o=1470 \text{ kg.m}^{-3}$ $\rho_w=1000 \text{ kg.m}^{-3}$ $C_{woeq}=0.01$
η	0.304	0.3	5	

All the experiments were carried out with constant filtration velocity of one meter per day ($V_L = 1m/day$) and with constant dispersion coefficient $D_w = 10^{-9} \text{ m}^2/s$. Figures 6 and 7 show that the fields obtained are characteristic of the local equilibrium situations. Figure-8 illustrates the temporal evolution of the concentration C_{wo} ($x=x_L, t$)/ C_{woeq} at the exit of the source zone. It is noticed that this evolution agrees with the predicted one by the theoretical considerations.

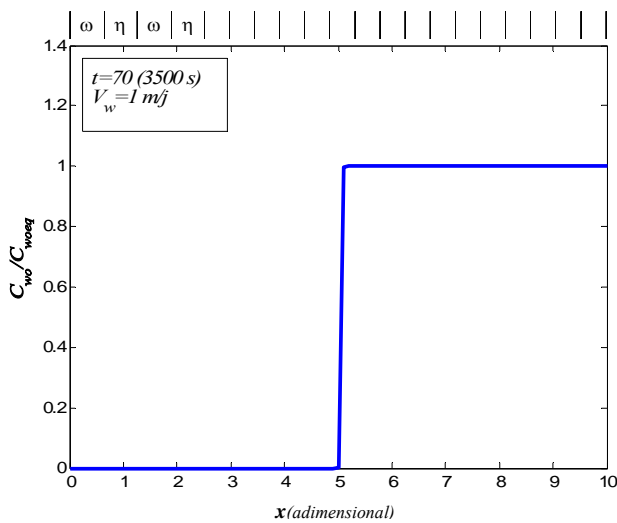


Figure-6. Concentration fields C_{wo}/C_{woeq} (in mass fraction) at $t=3500s$ and $V_w = 1m/day$ at Darcy-scale.

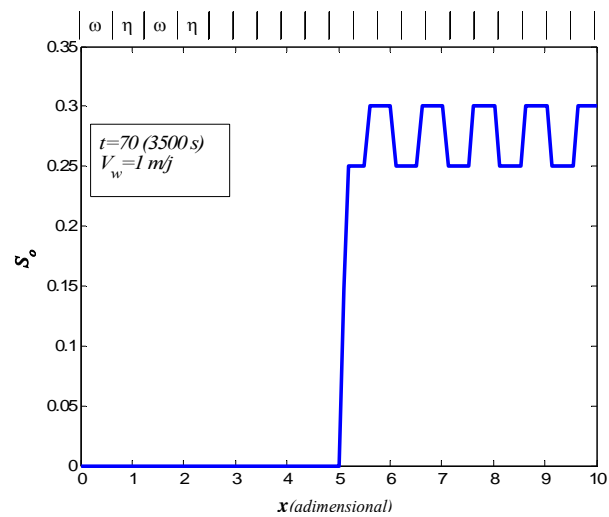


Figure-7. Saturation fields S_o at $t=3500s$ and $V_w = 1m/day$ at Darcy-scale.

The results presented below relate to the TCE dissolution in water in the same stratified porous medium as previously. Here, the Damköhler number (mass exchange coefficient) considered is however small in front of the unit in each stratum. The unit cell is made of two strata L_ω and L_η . The length ($L_\omega + L_\eta$) of the unit cell is 0.02 m.

Table-2. Data for the experiments relating to the local non equilibrium assumption.

Stratum	Porosity (ϵ)	TCE residual saturation (S_{or})	Mass transfer coefficient α (s^{-1})	Physical properties of the phases
ω	0.38	0.25	0.01	As in Table-1
η	0.304	0.3	0.02	

Figures 9 and 10 present respectively the concentration fields and saturation fields at time $t = 3500s$ for a simulation corresponding to the parameters contained in Table-2. The experiments were carried out for a

constant filtration velocity $V_w = 0.0004 \text{ m/s}$, and constant dispersion coefficient $D_w = 10^{-9} \text{ m}^2/s$

These results show that the Darcy scale fields can particularly have a space and temporal evolution rather



complex for the saturation when the velocity of flow in the porous medium is enough high. Figure-11 presents the evolution of the concentration as function of time at the exit of the domain of study. This result is in agreement with the theoretical predictions (Cf. § 2). It is noted in particular that in the case of a small mass exchange coefficient, the dissolution front is spread out over several unit cells.

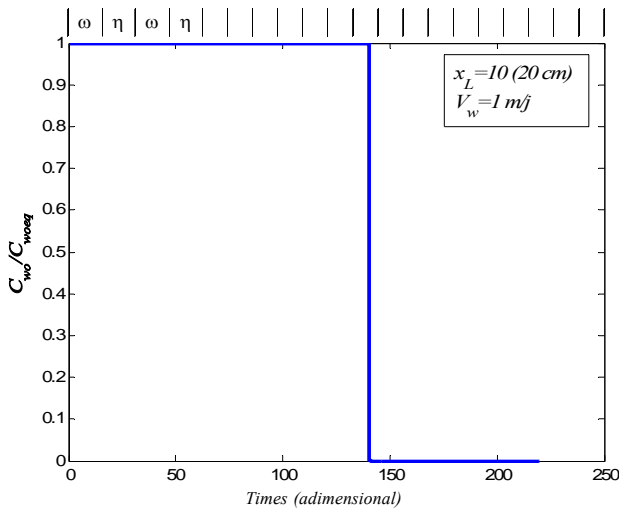


Figure-8. Concentration C_{wo}/C_{woeq} (in mass fraction) at $x=x_L$ (local equilibrium case).

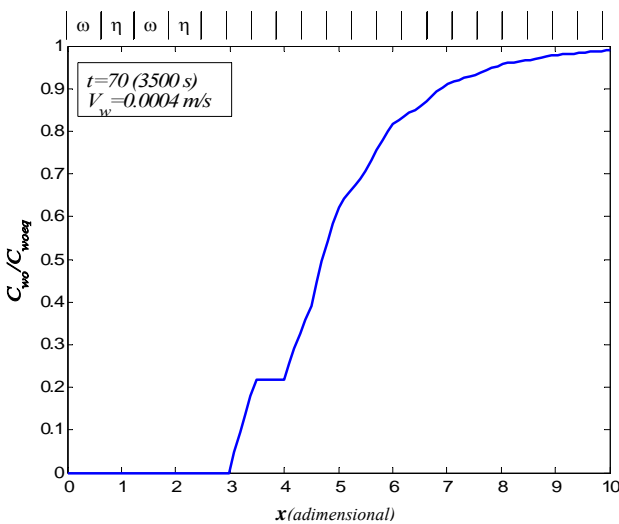


Figure-9. Concentration fields C_{wo}/C_{woeq} (in mass fraction) at $t=3500s$ at Darcy-scale.

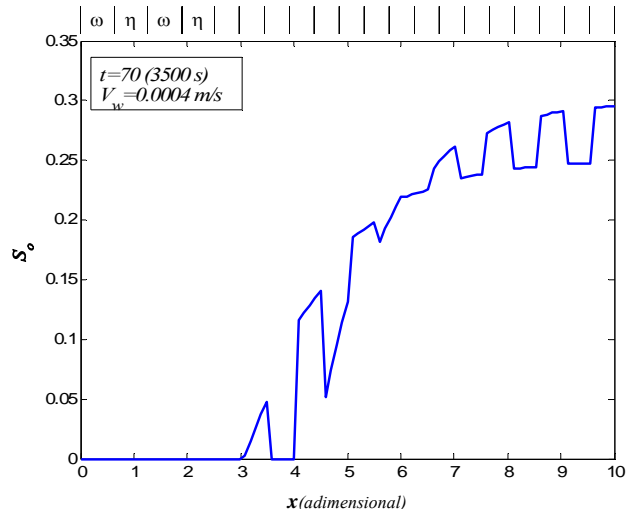


Figure-10. Champs de saturation fields S_0 at $t=3500s$ at Darcy-scale.

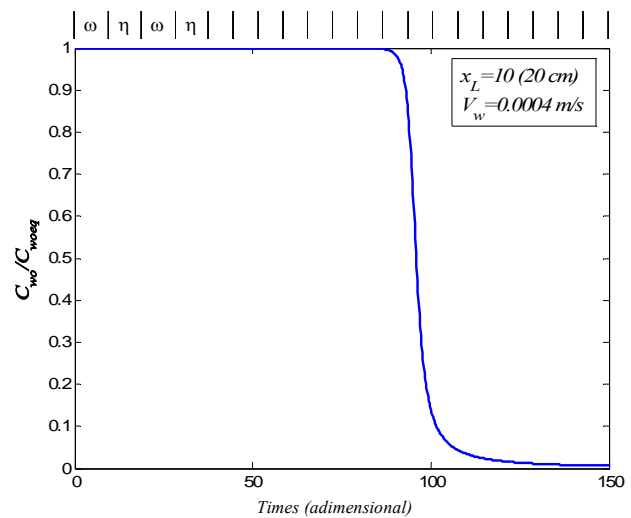


Figure-11. Concentration C_{wo}/C_{woeq} (in mass fraction) at $x=x_L$ local non equilibrium case.

4.2 Two-dimensional experiments (2D)

The modeling of dissolution in a domain contaminated by pollutant (here TCE) was carried out using the built program DISSOL 2D which takes into account three mechanisms: convection, hydrodynamic dispersion and the dissolution modeled by a coefficient of mass exchange. Velocity and pressure which reign in the medium during dissolution are determined by the resolution of the Darcy law (Eq. 12) supplemented by the continuity equation (Eq. 13) of the liquid phase in flow. Two water injections were carried out with pressure at entry of the domain (of size 10m x 0, 5 m): $P_{entry} = 10^5$ Pa and 2×10^5 Pa. The first pressure at entry generates flow velocities of 1.1×10^{-6} m/s in the less permeable stratum and of 8.7×10^{-6} m/s in the more permeable. When the injection pressure doubles, the flow velocities rates doubles. The pressure at exit P_{exit} is zero in both cases. The results presented below concern a medium initially



polluted at residual saturation with TCE (Figure-5). A water injection at $x = 0$, under a constant pressure starts the phenomenon of dissolution of the pollutant in this water. The boundary conditions of the problem are those described by Eqs. (15) to (17). At the domain entry, we impose the pressure and the concentration $C(0, y, t) = 0$ (in mass fraction). Initially, the domain is at residual saturation in each stratum as indicated in Table-3. The numerical simulations were carried out with the data contained in Table-3. The permeability relating to water is calculated using the correlation of Corey, $k_{rw}(S_o) = (1 - S_o)^2$.

Figures 12 to 17 respectively illustrate the evolution of the concentration fields and those of saturation at various times. For the values of the parameters selected, one notes that, as long as the

dissolution front did not leave the polluted zone, the concentration downstream from, of this zone remains equal to the equilibrium concentration. Moreover, water at weak concentration in pollutant advances in front of the dissolution front in the strata removed in pollutant and contributes to decrease the concentration in pollutant. It is noticed moreover that the process of dissolution is very long. Indeed, for examples considered here, it is after 1157.4 days that the pollutant starts to disappear from the domain polluted in the first stratum. Lastly, the two experiments cases treated enable us to observe that dissolution is significant when the velocity of flow in the porous medium is high and this is particularly marked in the stratum with high permeability (Figures 12 to 17).

Table-3. Data for the experiments relating to the local non equilibrium assumption.

Stratum	Porosity (ϵ)	TCE residual saturation (S_{or})	Permeability k (m^2)	Mass transfer coefficient α (s^{-1})	Physical properties of the phases
Ω	0.38	0.1	10^{-12}	5×10^{-5}	$P_o = 1470 \text{ kg. m}^{-3}$ $P_w = 1000 \text{ kg. m}^{-3}$ $C_{woeq} = 0.001$ $\mu_w = 10^{-3} \text{ kg. m}^{-1} \cdot \text{s}^{-1}$ $D_w = 10^{-9} \text{ m}^2 \cdot \text{s}^{-1}$
H	0.3	0.2	2×10^{-13}	5×10^{-5}	

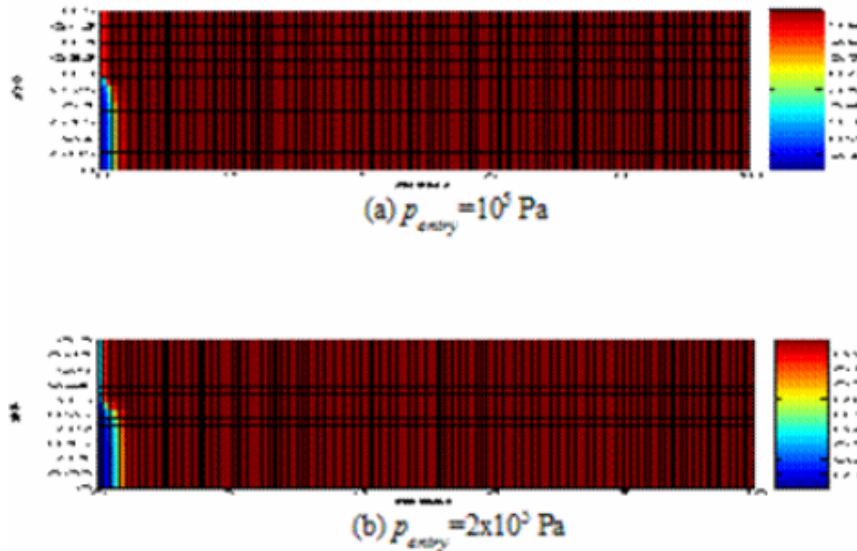


Figure-12. Concentration fields C_{w0}/C_{woeq} (in mass fraction) for $t=10^6 \text{ s} = 11.75 \text{ days}$, $\epsilon_o = .38$, $\epsilon_{\eta} = .3$, $k_o/k_{\eta} = 5$.

The concentration downstream from the source zone at point ($x=10 \text{ m}$, $y=0.5 \text{ m}$) takes a similar form to that predicted theoretically. On Figure-18, this form is characterized by: (i) a plate at equilibrium concentration, the source zone is sufficiently long (10 m in the direction of the flow), this is why the concentration is equal to equilibrium concentration during a certain time; (ii) a local non equilibrium situation illustrated by the reduction in

concentration (smaller than equilibrium concentration), to reach zero when the totality of the pollutant phase was dissolved. It is noted that the duration of disappearance of the source zone, by complete dissolution, is very long.

Concerning saturation downstream from the source zone at the same point ($x=10 \text{ m}$, $y=0.5 \text{ m}$), the behavior describes for the concentration is also observed in Figure-19.

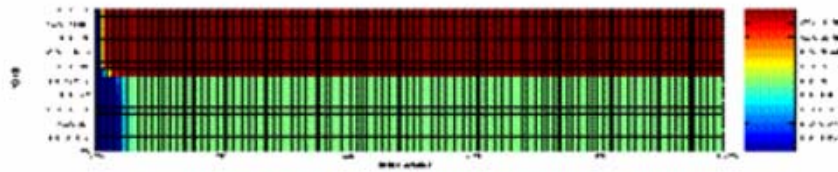
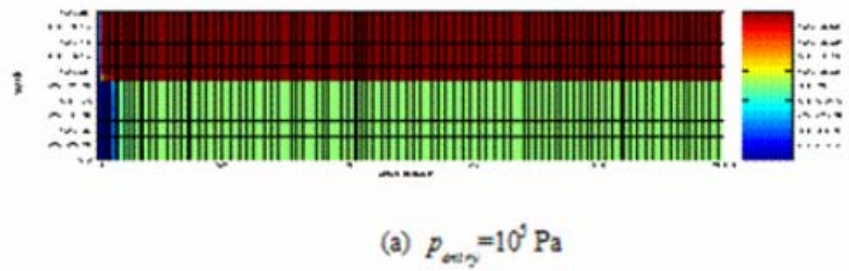


Figure-13. Saturation fields S_o for $t=10^6 \text{ s}=11.75 \text{ days}$ $\epsilon_o=.38$, $\epsilon_\eta=.3$, $k_o/k_\eta=5$.

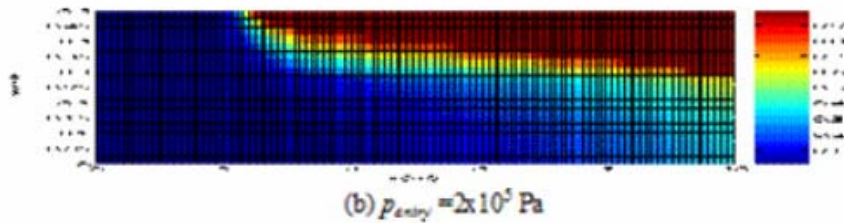
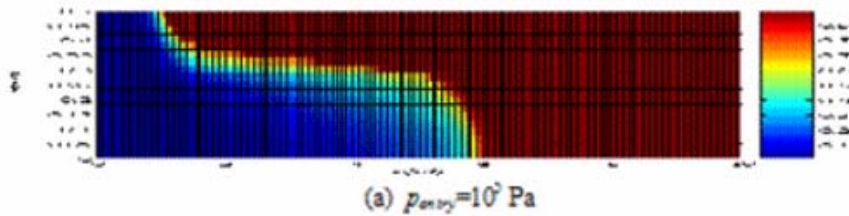


Figure-14. Concentration fields C_{w0}/C_{w0eq} (in mass fraction) for $t=5 \times 10^7 \text{ s}=578.70 \text{ days}$ $\epsilon_o=.38$, $\epsilon_\eta=.3$, $k_o/k_\eta=5$.

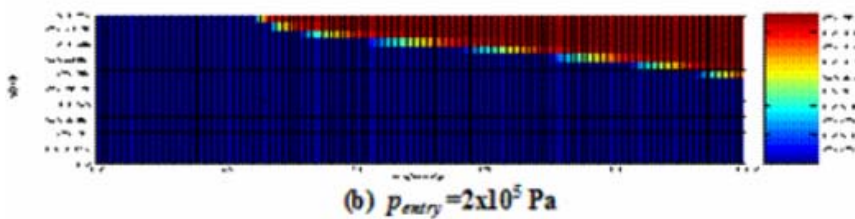
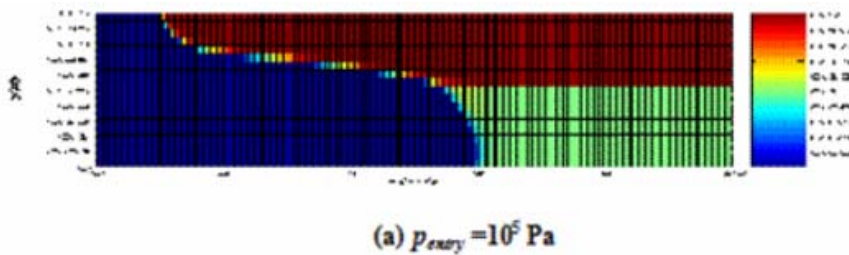


Figure-15. Saturation fields S_o for $t=5 \times 10^7 \text{ s}=578.70 \text{ days}$ $\epsilon_o=.38$, $\epsilon_\eta=.3$, $k_o/k_\eta=5$.

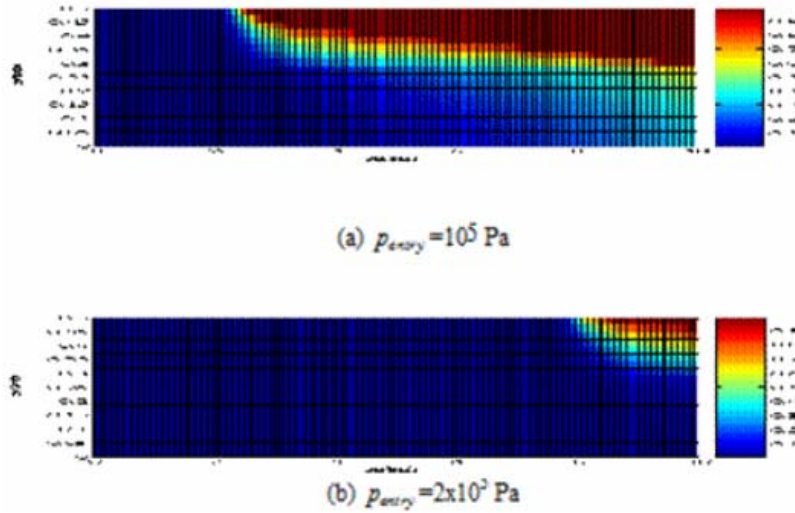


Figure-16. Concentration fields C_{wo}/C_{woeq} (in mass fraction) for $t=10^8$ s=1157.4 days $\epsilon_o=.38$, $\epsilon_\eta=.3$, $k_o/k_\eta=5$.

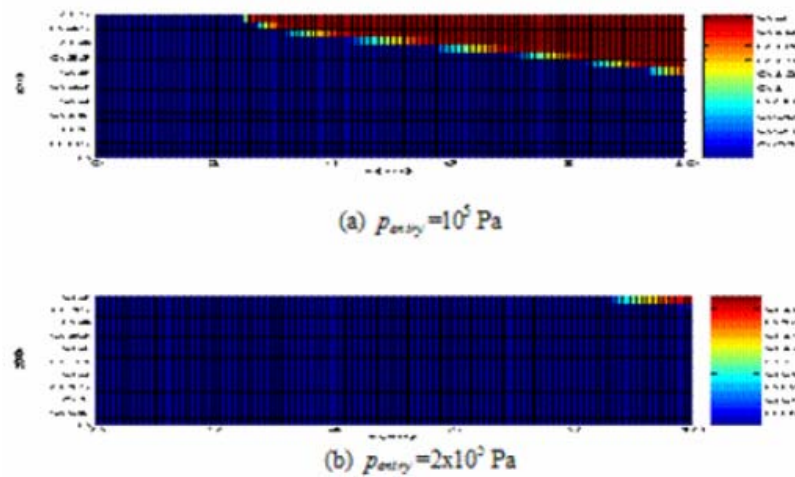


Figure-17. Saturation fields S_o for $t=10^8$ s=1157.4 days $\epsilon_o=.38$, $\epsilon_\eta=.3$, $k_o/k_\eta=5$.

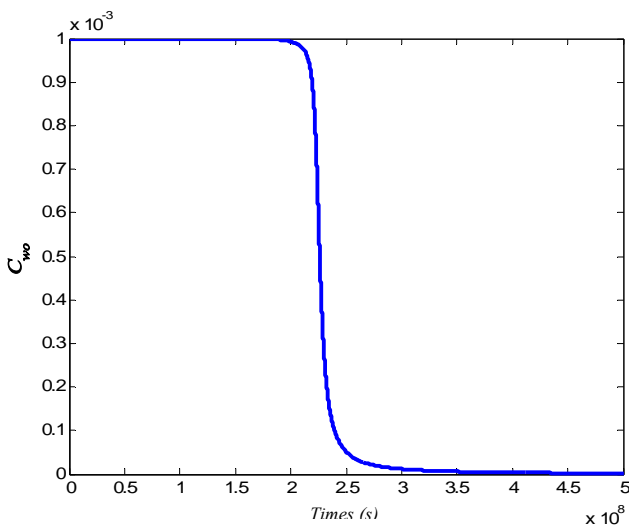


Figure-18. Concentration C_{wo} (in mass fraction) with the downstream of the source zone as function of time ($x=10$ m and $y=0.5$ m).

4.3 Heterogeneity effects on dissolution

We analyze in this section, the numerical simulation results obtained using the program DISSOL 2D. To appreciate the heterogeneity effects on dissolution on this strongly heterogeneous scale, this analysis relates to the pollutant saturation effects on dissolution. Porosity and permeability constitute the essential geometrical characteristics of the definition of the heterogeneity of media considered in this study. In the stratified porous medium considered, the form of the source zone is a significant data in the forecast of the dissolution of pollutants (Bradford *et al.*, 2003; Soga *et al.*, 2004, Côme *et al.*, 2005). We present hereafter, the experiment results simulating the saturation effects on the dissolution behavior at Darcy scale using the model developed in this study. The initial data used in the various experiments are available in Table-3.

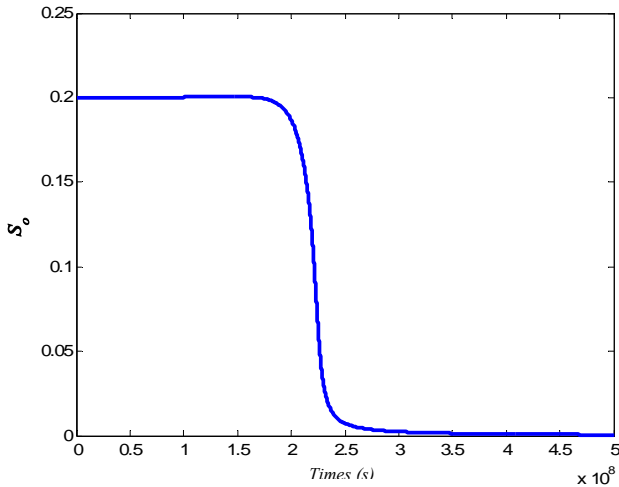


Figure-19. Saturation S_o with the downstream of the source area as function of time ($x=10$ m et $y=0.5$ m).

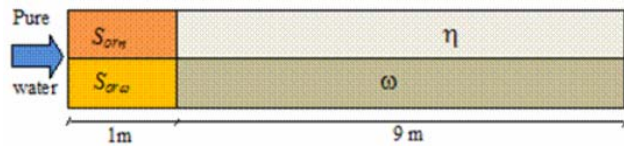


Figure-20. Initial saturation data.

The situation considered in the model is that schematized in Figure-20. We consider a stratified medium, initially polluted on one meter (source zone) by a residual saturation phase. Pure water which is injected in the medium charges in pollutant on totality of the section. According to the data of Table-3, the source zone is

modeled by a local non equilibrium model. Several numerical experiments ($t = 10^6$ s, 10^7 s, 5×10^7 s) were made on the situation described above. We show in the Figures hereafter the results obtained. The evolution of the saturation fields (Figure-21), show that a dissolution front progresses most fastly in the stratum with higher relative permeability to water (here the coefficient of mass transfer was selected the same one in the two layers). When the fast front reach the end of the source zone, the slow front continues its progression, with however a light deceleration due to the mechanism of deviation of the zone with low permeability. Possibly, the totality of the source zone can dissolve. Taking into account the reduction of velocity of the dissolution front due to deviation, and to the reduction of mass transfer coefficient, the final process can last a very long time.

However, one observes with regard to the concentration fields evolution (Figure-22) that downstream from the dissolution front, the concentration is equal to the equilibrium concentration and that upstream from this front, water with weak concentration in pollutant gradually advances, this situation remains as much as the dissolution front did not reach the end of the source zone. This process, after a relatively short time compared to the total time of dissolution of the source zone leads to a new mode of very slow evolution of the concentration, marked by a level of concentration at exit weaker than the equilibrium concentration because of the mixture of low polluted water passing by the completely dissolved stratum with strong polluted water of the stratum still contaminated.

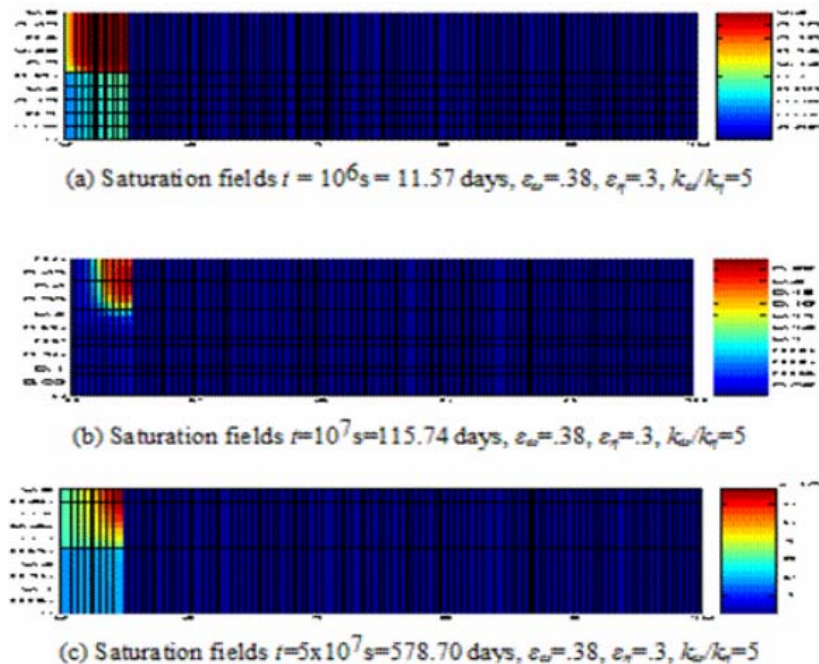


Figure-21. Saturation fields.

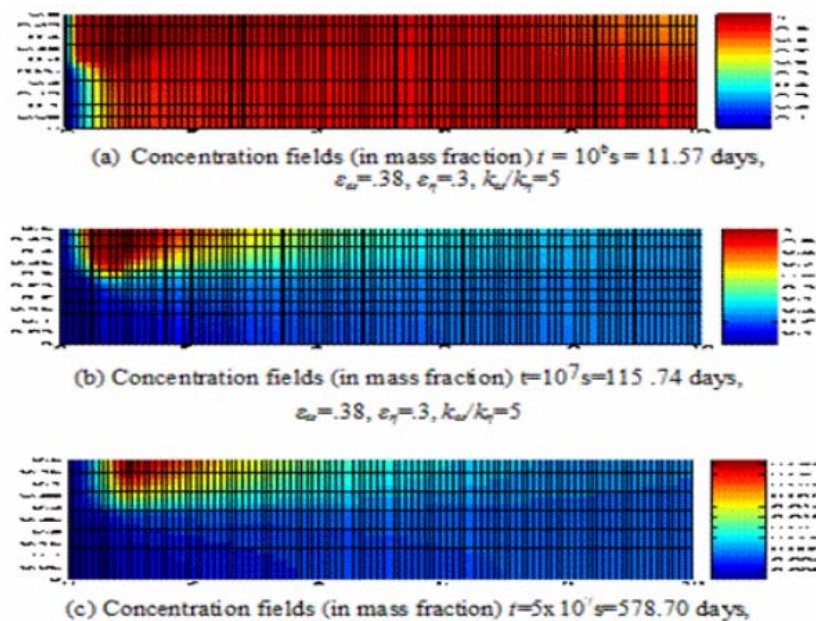


Figure-22. Concentration fields.

5. CONCLUSIONS

The model developed made it possible to simulate the dissolution of pollutants in heterogeneous porous media on the basis of simple 1D and 2D at Darcy scale. Two limit cases were studied: (i) local equilibrium case and (ii) local non equilibrium case. The model is based on an implicit numerical resolution of nonlinear partial differential equations. The numerical tests emphasize particularly the evolution of the exchange zone between the organic and water phases. Heterogeneity effects of the medium and heterogeneity effects of initial saturations induced by hydrodynamic instabilities or particular conditions of installation were characterized in agreement with the theoretical predictions, validating consequently the model built. Some experiments on the influence of heterogeneities on the model were carried out. They made it possible to highlight that dissolution is controlled by the medium heterogeneity coming from the geological and petrophysical characteristics of the matrix, like by the quantity of residual pollutant contained in poral space and its installation in the medium. These situations are of local non equilibrium type, because there exists in the porous medium, the saturated zones where mechanisms of exchange between water and the polluting phase occur, and where the concentration is far away from the equilibrium concentration.

In prospects, 3D-simulations on more complex heterogeneous media associating more complex numerical methods validated by experimental studies could constitute the continuation of this work.

ACKNOWLEDGEMENTS

We wish to thank Mr. Michel QUINTARD, Director of Research at CNRS, Toulouse Institute of Fluids Mechanics, France, for his guidance and financial support in the success of this work.

REFERENCES

- Bear J. 1961. On the tensor form of dispersion in porous media. *J. Geophys. Res.* 66: 1185-1197.
- Bear J. 1972. *Dynamics of Fluids in Porous Media*. American Elsevier, New-York, 764 pages.
- Bradford S. A., Rathfelder K. M., Lang J., Abriola L. M. 2003. Entrapment and dissolution of DNAPLs in heterogeneous porous media. *Journal of Contaminant Hydrology*. 67(1-4): 133-157.
- Brenner H. 1980. Dispersion resulting from flow through spatially periodic porous media. *Trans R Soc.* 297(1430): 81-133.
- Cherblanc F. Ahmadi A., Quintard M. 2003. Two-medium description of dispersion in heterogeneous porous media: calculation of macroscopic properties, *Water Resources Research*. Vol. 39, No. 6, p. 1154. DOI: 10.1029/2002WR001559.
- Côme J. M., Burghoffer P., Danquigny C., Emoney-Gauthier A., Getto D., Haeseler F., Kaskassian S., Le Roux F., Quintard M., Razakarisoa O. and Schaefer G. 2005. CIDISIR, Quantification des cinétiques de dissolution sur sites réels - hydrocarbures pétroliers et dérivés halogénés, Guide méthodologique, Programme RITEAU/ Ministère de la recherche. p. 72.
- Danckwerts P. 1953. Continuous flow systems distribution of residence times. *Chem. Engng. Sci.* 2: 1-13.
- Edwards M.G., Rogers C.F. 1994. A flux continuous scheme for the full tensor pressure equation. *Proc.* 4th



- European Conference on the Mathematics of oil Recovery D. pp. 1-8.
- Illangasekare T. H., Armbruster E. J., Yates D. N. 1995. Non-aqueous phase fluids in heterogeneous aquifers: An experimental study. *Journal of Environmental Eng.* 121(8): 571-579.
- Illangasekare T. H., Ramsey J. L., Jensen K. H., Butts M. 1995. Experimental study of movement and distribution of dense organic contaminants in heterogeneous aquifers: An experimental study. *Journal of Contaminant Hydrology.* 20(1): 1-25.
- Lowry M. I., Miller C. T. 1994. Pore-scale modelling of nonwetting-phase residual in porous media. *Center for Multiphase Research News.* 1(1): 7-9.
- Lowry M. I., Miller C. T. 1995. Pore-scale modeling of nonwetting-phase residual in porous media. *Water Resources Research.* 31(3): 455-473.
- Mabiala B. 2009. modélisation des pollutions des sols par des composés hydrocarbonés: effets des hétérogénéités sur la dissolution. Thèse de Doctorat d'état, Université Marien Ngouabi, Brazzaville. p. 160.
- Mabiala B., Tathy C. and Quintard M. 2004. Dissolution d'une phase polluante hydrocarbonée en milieu poreux hétérogène. *URED, revue pluridisciplinaire de l'U. G. B de St-Louis du Sénégal.* 10: 23-33.
- Mabiala B., Tathy C. and Quintard M. 2003. Napl Dissolution in Heterogeneous Systems: Large-Scale Analysis for Stratified System. In: HEFAT 2003, 2nd International Conference on Heat Transfer, Fluid Mechanics and Thermodynamics. Victoria Falls, Zambia.
- Mayer A. S. and Miller C. T. 1996. The influence of mass transfer characteristics and porous media heterogeneity on nonaqueous phase dissolution. *Water Resources Research.* 32: 1551-1561.
- Mayer A., Endres K. L. 2007. Simultaneous optimization of dense non-aqueous phase liquid (DNAPL) source and contaminant plume remediation. *Journal of Contaminant Hydrology.* 91: 288-311.
- Parker J. C., Katyal A.K., Kaluarachchi J.J., Lenhard R.J., Johnson T.J., Jayaraman K. *et al.*, 1991. Modelling multiphase organic chemical transport in soils and ground water, Technical report Environmental Protection Agency Project Robert S., Kerr. Environmental Research Laboratory, series volume ID: EPA/600/2-91/042: 1991.
- Park E., Parker J.C. 2005. Evaluation of an upscaled model for DNAPL dissolution kinetics in heterogeneous aquifers. *Advances in Water Resources.* 28: 1280-1291.
- Powers S. E., Abriola L. M., Weber W.J. Jr. 1992. An experimental investigation of nonaqueous phase liquid dissolution in saturated subsurface systems: Steady state mass transfer rates. *Water Resources Research.* 28(10): 2691-2705.
- Quintard M. and Whitaker S. 1994. Convection, dispersion, and interfacial transport of contaminants: Homogeneous porous media. *Advances in Water Resources.* 17: 221-239.
- Radilla Zuazo G. 1997. Contamination des eaux souterraines par les hydrocarbures: expériences en laboratoire, modélisation, interprétation et résolution des problèmes inverses pour l'estimation des propriétés de transport. Th. Doctorat, ENSAM Bordeaux. p. 210.
- Saenton S., Illangasekare T. H., Soga K., Saba T. A. 2002. Effects of source zone heterogeneity on surfactant-enhanced NAPL dissolution and resulting remediation end-points. *Journal of Contaminant Hydrology.* 59: 27-44.
- Soga K., Page J. W. E., Illangasekare T. H. 2004. A review of NAPL source zone remediation efficiency and the mass flux approach. *Journal of Hazardous Materials.* 110: 13-27.
- Urgelli D. 1998. Prise en compte des hétérogénéités par prise de moyenne des transmissivités sur maillage adaptés en simulation de réservoir, Thèse de 3^e cycle, Université de Provence (Aix-Marseille I).
- Yra A. 2006. Dispersion active en milieux poreux hétérogènes contaminés par des produits hydrocarbonés. Thèse de Doctorat de l'Université de Bordeaux I. p. 123.
- Zhang Changyong, Hongkyu Yoon, Werth Charles J., Valocchi Albert J., Basu Nandita B., Jawitz James W. 2008. Evaluation of simplified mass transfer models to simulate the impacts of source zone architecture on nonaqueous phase liquid dissolution in heterogeneous porous media. *Journal of Contaminant Hydrology.* 102: 49-60.