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## A Mathematical Model for Nanoparticles Transport within Two-phase Flow in Heterogeneous Porous media

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**Abstract — A mathematical model for nanoparticles transport within two-phase flow in heterogeneous porous media** — *Transport of nanoparticles in porous media is a growing concern among scientific research and technological development, such as in the preservation of groundwater quality, pollution control and remediation, and enhancement of oil recovery. Indeed, recent advances in the later field suggested significant improvements in the recovered volumes by injecting hydrophobic nanoparticles which enhance or reverse the initial reservoir wettability favoring an increase in the relative permeability of the oil phase and the capillary pressure gap between phase pressures. A new mathematical model, based on previous work by Sbai and Azaroual [Adv. Water Resour. 34(1), 62-68, 2011] is developed which couples the incompressible two-phase fluid flow reservoir equations at the macroscopic level to equations of nanoparticles transport at a smaller, but still macroscopic, secondary scale. The latter accounts for pore scale processes of colloidal and hydrodynamic release from pore surfaces, deposition in pore surfaces and possibly clogging the pore throats. The mechanism of particles interphase transfer is accounted for based on the wettability of nanoparticles surfaces. Nanoparticles are divided among several classes based on their size and wettability properties. Each class is assumed to have a constant colloidal and hydrodynamic detachment rates taken as first-order kinetics, a constant surface deposition rate in pore walls of the same wettability according to filtration theory, and a constant mass capture rate at pore throats by blocking and bridging. For each nanoparticle class a mass convection-diffusion equation with a total rate of all related mechanisms is solved numerically with a finite volume method. Change of absolute permeability is correlated to porosity change resulting from mass distributions of flowing, surface, and pore-throat deposited nanoparticles. Relative permeability and capillary pressure functions are optionally allowed to be time-dependent as a function of the mass fraction of deposited nanoparticles of the same wettability. Many multi-dimensional applications of the presented model are given for performance evaluation. The newly developed model has many applications and opens interesting perspectives for environmental remediation, well injectivity design, re-engineered fluids injection, and CO<sub>2</sub> sequestration in deep geological formations. Finally, a nested multiscale finite element method is applied to this problem where three hierarchical scales are considered for the resolution of the global pressure, fluid saturation, and nanoparticles transport equations. An important speedup is achieved by this procedure allowing for realistic engineering grade applications to be tackled efficiently.*

### INTRODUCTION

Transport of nanoparticles (NP's) in porous media is a growing concern among scientific research and technological development. In earth sciences, several emerging applications have been reported in several fields, such as in the preservation of groundwater quality (Cullen et al., 2010), pollution control and remediation, fines migration control (Huang et al., 2008), and enhancement of oil recovery (Ju et al, 2006; Fletcher and Davies, 2010). Recent advances in the later field suggested significant improvements in the recovered volumes by injecting hydrophobic nanoparticles which enhance or reverse the initial reservoir wettability favoring an increase in the relative permeability of the oil phase and the capillary pressure drop between phase

pressures (Ju and Fan, 2006; Onyekonwu and Ogolo, 2010). A new mathematical model have been developed for the purpose of numerical interpretation and analysis of laboratory experiments of flooding petrographic cores with different nanofluids. The numerical model is elaborated for prediction of micro- and nano-particle mobilization, migration, and capture in porous media systems at the aquifers and reservoirs scale. This extended abstract highlights several key features of our numerical engine including physical and chemical processes taken into account. The developed framework involves solving a set of partial differential equations (PDE's) coupled to particle kinetics in particular. Resolution of this system is notoriously a computationally challenging task. We highlight the main numerical methods used for spatial discretization of

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from pore to field scale. *Pore2Field*  
16-18 November 2011, IFP Energies nouvelles (France)**

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these equations and describe our current efforts to improve computational efficiency and discretization on flexible and fully unstructured grids to be able to discretize the underlying equations on different subscales. Next, a couple of example applications are shortly described and commented. The first example involves the mobilization, migration, and capture in pore throats of endogenous micron size particles in the context of CO<sub>2</sub> injection in sandstones. The impact of precipitates from strong CO<sub>2</sub>-Water-Rock interactions on the injectivity while considering fine scale heterogeneity of the reservoir is analyzed. The second example describes a simulation of enhanced oil recovery in a heterogeneous reservoir while flooding with hydrophilic nanoparticles. It is shown that the numerical model is able to predict an improvement in the recovered volumes due to surface wettability effects, while noting a slight decrease in the absolute permeability. This suggests development of a modeling-optimization procedure for an optimal oil recovery while maintaining the well injectivity.

## **1 PHYSICAL PROCESSES AND CONCEPTUAL MODEL**

Sbai and Azaroual (2011) describe in detail the conceptual model for particulate transport processes in two-phase flow systems. Their work has involved model verification on previously published experimental data. The developed numerical engine was used equally to predict mobilization, migration, and capture of CO<sub>2</sub>-wet particles in homogeneous and heterogeneous permeability fields to assess the impact of fine scale geological heterogeneity on the injectivity during CO<sub>2</sub> storage. The mathematical model is based on a coupling between incompressible two-phase flow equations for reservoir simulation and a deep bed filtration model, which considers migration in each fluid phase and interphase particulate mass transfer. This set of governing equations is complemented by an optional advection-dispersion solute transport equation in the aqueous phase to calculate the spatial distribution of salinity, which could trigger the mobility of in-situ colloidal fines migrating in the aqueous phase. Pore scale particulate transport processes are described by a set of coupled convection-diffusion equations for each subset of NP's. The couplings occur only over quasi-linear rate laws for basic processes of release from pores bodies, mobility in fluid phases, and capture in pores throats. Quantitative assessment of the permeability decline is strongly dependent on the accuracy of experimental data on permeability-porosity

relationships as was discussed and shown previously (Sbai and Azaroual, 2011).

## **2 NUMERICAL METHODS**

A fully coupled solution of the governing equations would be a daunting task. We choose instead a sequential formulation, which has the add-on advantage to select among other on-the-shelf reservoir codes (i.e. a three-phase flow Black oil or compositional model). The incompressible two-phase flow model is based on a fully implicit solution of the global pressure equation using the standard TPFA first-order upwind finite-volume scheme. Coupling between the global pressure and phase saturation equations is performed with an IMPES or IMPSAT schemes favoring sharp fronts localization and computational efficiency, respectively. The intermediate algebraic nonlinear equations related to the saturation equation are solved with a Newton-Raphson adaptive procedure combined with a line-search technique. After recovery of the phase fluid velocities in an intermediate step, the convection-diffusion-reaction like equations for a set of involved particles are solved by operator splitting techniques. The chief advantage of such techniques being use of highly specialized solvers for each module. Particle kinetics are integrated with DASPK computer package. Particle rates are estimated in a post-processing step. Finally, at the end of each time step porosity and permeability changes are evaluated based on the mass of particles being deposited and/or mobilized in pore surfaces and pore throats. We consider a hierarchical time stepping loop involving the pressure, phase saturation, and particles concentrations variables, respectively.

## **3 APPLICATIONS**

### **3.1 Modeling formation damage by CO<sub>2</sub> injection in a heterogeneous subsurface aquifer**

CO<sub>2</sub> injection for sequestration projects in sandstone formations involves mobilization, migration, and capture of endogenous and exogenous fine particles in porous media. Examples of endogenous particles are salt deposits, and precipitating secondary mineral phases due to strong geochemical reactivity with the primary minerals. Examples of endogenous particles are mobilized metal oxides from corroded metal pipes or those present in the gas stream as a result of the CO<sub>2</sub>

**Flows and mechanics in natural porous media  
from pore to field scale. Pore2Field  
16-18 November 2011, IFP Energies nouvelles (France)**

capture process together with CO<sub>2</sub> that cannot be completely avoided for economic reasons.

In this application we consider a two-dimensional permeability map extracted from the full permeability field used by the SPE-10 2<sup>nd</sup> comparative solution project. This map corresponds to the 36<sup>th</sup> layer in the Upper-Ness formation sequence where permeability is fluvial and could be characterized as a high-contrast heterogeneous porous medium. The selected layer is comprised of 60 x 200 (13,200) cells. The porosity is strongly correlated with the horizontal permeability and contains about 2.5% null values. To avoid division by zeros in these cells, we simply replace the null values by a threshold minimal value. The oil phase is extracted from four production wells at the domain corners with an equal flow rate and CO<sub>2</sub> is injected into the formation from an injection well in the center.

Figure 1 shows simulation results after 30 days of CO<sub>2</sub> injection. The injectivity decline is slightly higher due to an already well-established high-permeability channel and a high contrast with its surrounding. Here the particle capture mechanisms are exclusively controlled by the medium heterogeneity and not by the hydraulic interactions. Indeed the narrow channel volume intercepts most of the particles at the pore surfaces and pore throats. Additionally, we can see that injectivity loss distribution in the reservoir is not gradually decreasing radially from the injection well. It is not anymore restricted close to the injector and could be reached in other parts of the reservoir, as in the most permeable channel composed from fluvial sediments, or in fragmented sets of spots at the vicinity of some production wells.

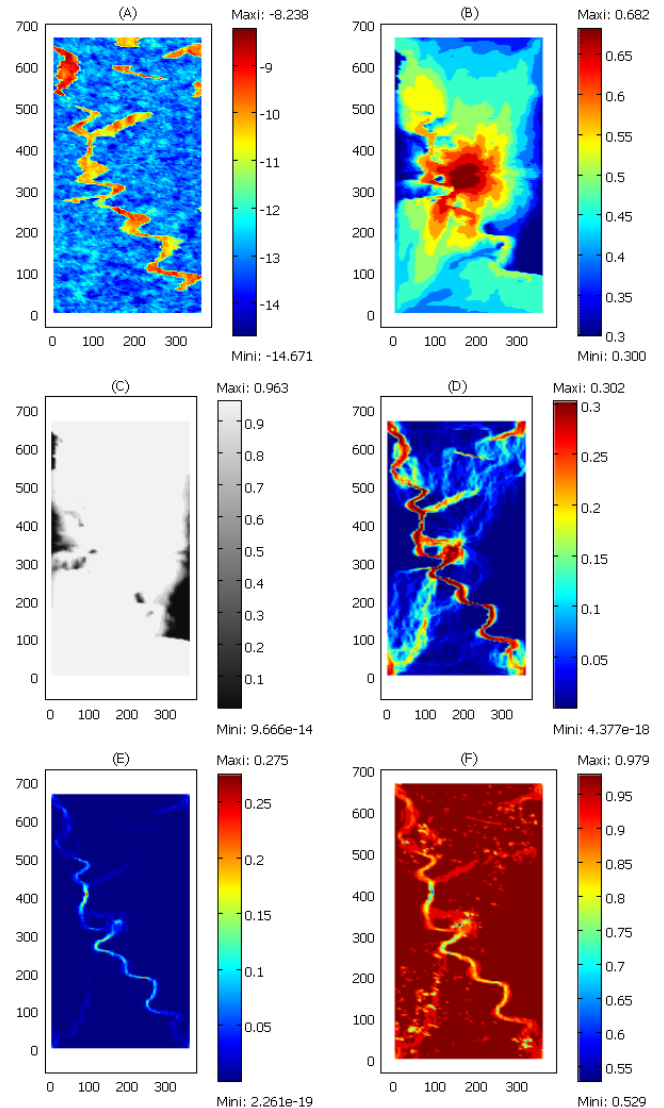


Figure 1 – Simulation results after 1 month of CO<sub>2</sub> injection in a five-spot well pattern showing spatial distribution of (A) the initial reservoir permeability (log<sub>10</sub>-scale) (B) CO<sub>2</sub> phase saturation, mass concentrations of (C) suspended particles, (D) deposited particles in pore surfaces, and in (E) pore throats, and (F) permeability reduction factor.

**3.2 Improving oil recovery in a permeable reservoir by injection of hydrophilic NP's**

Ju and Fun (2009) combine an experimental and a one-dimensional core-scale numerical modeling for flooding oil-wet rock with lipophobic and hydrophilic polysilicon nanoparticles (LHPN's). They noticed a change of the rock wettability, during the experiment, from oil-wet to

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from pore to field scale. *Pore2Field*  
16-18 November 2011, IFP Energies nouvelles (France)**

strongly water wet. The contact angle between the rock surface and the water phase decreased sharply from above to below  $90^\circ$ . Improvement in the recovered oil volume is due to two processes, (i) the change in the rock wettability, and (ii) the decrease of the interface tension when injecting a LHPN.

Here, we consider a two-dimensional domain having identical size and dimensions of the first example discussed above, but the permeability distribution is rather smooth and is extracted from the top layer of the SPE-10 2<sup>nd</sup> comparative solution project's full grid. We consider the same well pattern except for the total flow rate, which equals  $50 \text{ m}^3/\text{h}$ .

Figure 2 shows the water and oil cuts during the first 30 days of production from the upper right well. The numerical model clearly predicts an improved oil recovery when the reservoir is flooded with LHPN's, while noting a decrease in the mobility of the water phase and a relative increase in the oil mobility. This induces a higher residence time of the water-wet nanoparticles, and hence treatment with LHPN's shows a net decrease of the intrinsic permeability nearby the injection well. For a highly permeable reservoir, it is expected that such formation damage does not offset the benefit of reservoir treatment by LHPN's. However, for reservoirs with low initial permeability it is advised to perform such modelling coupled to an optimization procedure.

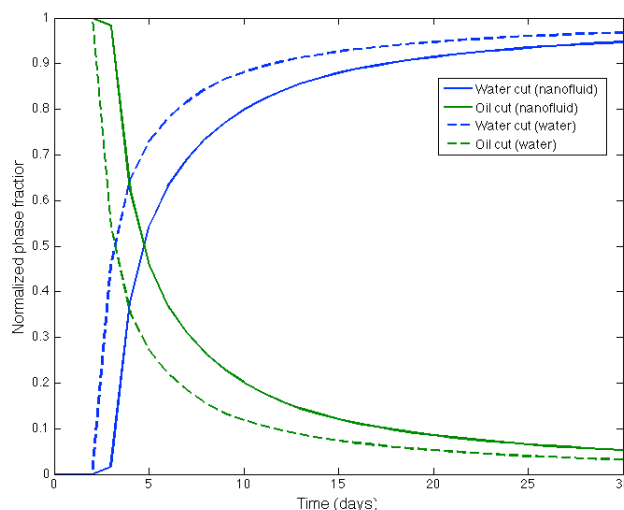


Figure 2 – Comparison between oil and water cuts obtained by injection of water with and without LHPN's. The curves indicate a net improvement in the net recovered oil volume.

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