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Modeling the evolution of spectral induced polarization during calcite precipitation on glass beads

Leroy Philippe
BRGM
Orléans, France
p.leroy@brgm.fr

Li Shuai
Imperial College
London, England
lshuai8002@yahoo.com

Jougnot Damien
CNRS, UMR 7619 METIS
Paris, France
damien.jougnot@upmc.fr

Revil André
CNRS, UMR 5275
Le Bourget du Lac, France
andre.revil@univ-smb.fr

Wu Yuxin
LBNL
Berkeley, USA.
ywu3@lbl.gov

SUMMARY

When pH and alkalinity increase, calcite frequently precipitates and hence modifies the petrophysical properties of porous media. The complex conductivity method can be used to directly monitor calcite precipitation in porous media because it is very sensitive to the evolution of the pore structure and its connectivity. We have developed a mechanistic grain polarization model considering the electrochemical polarization of the Stern layer surrounding calcite particles. This model depends on the surface charge density and mobility of the counter-ions in the Stern layer. Our induced polarization model predicts the evolution of the size of calcite particles, of the pore structure and connectivity during spectral induced polarization experiments of calcite precipitation on glass beads pack. Model predictions are in very good agreement with the complex conductivity measurements. During the first phase of calcite precipitation experiment, calcite crystals growth, and the inverted particle size distribution moves towards larger calcite particles. When calcite continues to precipitate and during pore clogging, inverted particle size distribution moves towards smaller particles because large particles do not polarize sufficiently. The pore clogging is also responsible for the decrease of the connectivity of the pores, which is observed through the increasing electrical formation factor of the porous medium.

Key words: calcite precipitation, complex conductivity, Stern layer, particle size, pore clogging.

INTRODUCTION

Calcite is one of the most abundant minerals in the earth crust and frequently precipitates when alkalinity and pH increase (Vancappellen *et al.*, 1993). Calcite precipitation modifies the rock porosity, and can have positive or harmful effects for the mechanical and transport properties of porous media. Calcite precipitation in porous media has broad applications in geotechnical engineering for soil strengthening (DeJong *et al.*, 2006) and in environmental studies for the sequestration of heavy metals (Sturchio *et al.*, 1997), radionuclides (Fujita *et al.*, 2004) and CO₂ in geological formations (Pruess *et al.*, 2003). However, calcite precipitation can also have undesirable effects such as the decrease of the efficiency and permeability of reactive barriers for the remediation of aquifers (Wilkin *et al.*, 2003).

Wu *et al.* (2010) performed complex conductivity measurements and modeling of calcite precipitation on glass beads packed column. From their imaginary part of complex

conductivity data, the evolution of calcite precipitation in porous media was clearly observed. The empirical Cole-Cole model (Cole and Cole, 1941) was used by Wu *et al.* (2010) to interpret the complex conductivity signature of calcite precipitation in glass beads. However, the lack of physical processes in the Cole-Cole model to interpret the complex conductivity data restricts the understanding of the effects of calcite precipitation on the evolution of the pore structure and connectivity in glass beads column. The induced polarization of calcite precipitates needs to be further clarified using a mechanistic complex conductivity model accounting for the EDL properties and the particle size distribution. In this study, a mechanistic model for the induced polarization of calcite is proposed, which depends on the surface charge density and ions mobility of the counter-ions in the Stern layer and on the particle size distribution. The predictions of the model are compared to the imaginary conductivity data of Wu *et al.* (2010), and the evolution of the pore structure during calcite precipitation in glass beads is estimated accordingly.

THEORETICAL BACKGROUND AND COMPARISON WITH EXPERIMENTAL DATA

We consider a porous medium containing particles, glass beads grains (of millimetric size) and calcite crystals (of micrometric size), and water (subscript “w”). The complex conductivity model is presented at Figure 1.

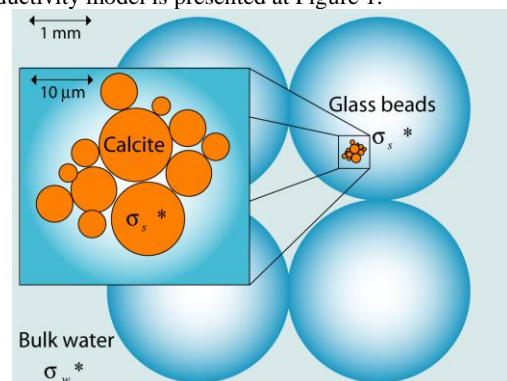


Figure 1. Sketch of the complex conductivity model of the porous medium.

Maxwell-Wagner polarization occurs at the boundary between the different phases (solid, water) possessing different electrical properties. The differential effective medium (DEM) theory (Sen *et al.*, 1981) is used to compute the electrical conductivity of the porous medium according to the conductivity of the particles and liquid. The complex surface conductivity of the particles of different sizes is calculated considering the superposition principle and using the particle

with the visual observations from SEM (scanning electron microscopy) images in the experiment (Wu *et al.*, 2010). The calcite particles increase approximately from less than 1 to 20 μm , as reported by Wu *et al.* (2010). During the first stage of calcite precipitation, the modeled volume of the pore water decreases due to calcite precipitation (Figure 6).

As the calcite precipitation experiment continued over 9 days, the clogging occurred in the sample holder. At the second stage, the formation factor of glass beads increases significantly as shown in Figure 6, from 6.08 to 7 (day 10), 11 (day 11) and 12 (day 12). The changes of the formation factor is due to the loss of connectivity of glass beads pores affected by the clogging even though the porosity of the sample (glass beads, porous medium) has a tiny change. The modeled particle size distribution obtained from the inverted imaginary conductivity spectra (quadrature conductivity) moves towards smaller particles as experiment continues (Figure 4b and Figure 5b). This could be explained by that the large calcite particles created during the clogging process do not play an important role in the complex conductivity spectra (they do not polarize sufficiently) and only smaller particles are the effective ones contributed to the complex surface conductivity. Calcite precipitation induces a smaller pore volume fraction, therefore, a slight increase of the formation factor F . The occurrence of pore clogging may explain the increase of the cement exponent from 1.5 to 2 for glass beads materials, which leads the formation factor F increasing from 6.08 to 12 under the same porosity.

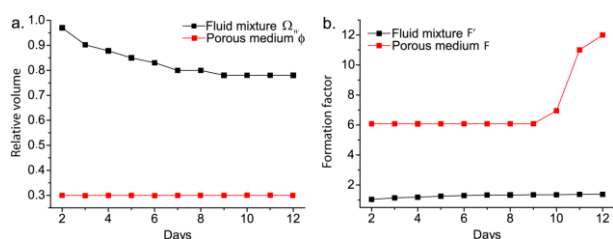


Figure 6. Computed relative volume of the fluid to the volume of the porous medium and relative volume of the bulk water to the volume of the fluid mixture and formation factors F and F' changes during the calcite precipitation experiment of Wu *et al.* (2010). The pore clogging happens at day 9, the formation factor of the porous medium (glass beads) changes dramatically.

CONCLUSIONS

A mechanistic complex conductivity model was used to interpret spectral induced polarization experiments of calcite precipitation on millimetric glass beads containing CaCl_2 and Na_2CO_3 aqueous electrolytes in equal concentration. The conductivity model considers the electrochemical polarization of the Stern layer surrounding calcite particles and depends on the surface site density and surface mobility of counter-ions in the Stern layer, which were kept constant during the simulation of the precipitation experiment. The particle size distribution, porosity and electrical formation factor evolution during the precipitation process were inverted from imaginary conductivity data.

Model predictions are in very good agreement with the measured imaginary conductivity spectra and the microscopy observations of the evolution of the pore structure and connectivity during calcite precipitation. The tangential mobility of the counter-ions in the Stern layer is found to be

similar to their mobility in bulk water. The kinetic of calcite precipitation in glass beads column is described by considering two different stages, one before the pores clogging where modeled particle size distribution moves to larger particles due to the growth of calcite crystals, and another during the pores clogging where only the smaller particles influence the polarization response. During the first stage of calcite precipitation, the electrical formation factor of glass beads remains constant and the modeled pore water volume decreases due to calcite precipitation. During the second stage of calcite precipitation, the electrical formation factor of glass beads increases considerably because of the loss of pores connectivity due to the clogging process and the modeled pore water volume remains constant. These observations can be explained by the aggregation of the calcite precipitates merging at the surface of glass beads, which can significantly alter the connectivity and current paths of the pore space of glass beads even though the total porosity remains nearly unchanged.

This study shows that spectral induced polarization can be an efficient and cost effective geophysical method to monitor non-invasively and continuously calcite precipitation in porous media because of its sensitivity to polarization processes occurring at the mineral/water interface. A mechanistic induced polarization model is also necessary to interpret induced polarization experiments in terms of evolution of particle size distribution, pores structure and connectivity during calcite precipitation.

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