Effect of the local clay distribution

on the effective elastic properties of shales

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Accepted for publication in Mechanics of Materials
Abstract

This paper revisits the “local porosity theory” developed by Hilfer to propose a “local clay theory” (LCT) that establishes a quantitative relationship between the effective elastic properties and clay distribution of shales. This approach is primarily based on a “local simplicity” assumption; under this assumption, the complexity of spatial clay distribution can be captured by two local functions, namely, the local clay fraction distribution $\mu$ and the local percolation probability $\lambda$ which are calculated by partitioning a mineral map. The local clay fraction distribution provides information about spatial fluctuations of clay fraction and the local percolation probability describes the spatial fluctuations in the clay connectivity.

This LCT was applied to a mineral map made from a Callovo-Oxfordian mudstone sample for which petrophysical data were available. This application demonstrated that the local functions $\mu$ and $\lambda$ can be reasonably approximated by a Gaussian and simple sigmoid function, respectively.

LCT was also used in a sensitivity analysis to evaluate the impact of the spatial fluctuations of the clay phase and clay connectivity on the effective elastic properties of shales. This impact has been investigated by comparing the effective properties obtained from LCT with those provided by a percolating inclusion-based model (a non-clay spherical grain embedded in a transverse isotropic clay matrix with neither spatial clay fluctuations nor spatial fluctuations in the clay connectivity). This inclusion-based model was built from a Differential Effective Medium scheme. The results of this sensitivity analysis indicated that spatial clay fluctuations have only a slight impact on the calculated effective properties. However, when the spatial fluctuations of clay connectivity were considered, the quantitative deviations between the effective properties inferred from LCT and those from the percolating inclusion-based model were large exceeding 50% in some cases.

Keywords: elastic properties, microstructure, shale, clay-rocks, clay minerals, effective medium approximations. Short title: shale microstructure and elastic properties.
1. Introduction

Clay minerals are the major components of shales which are the main geological caps to many hydrocarbon reservoirs (e.g., Mavko et al., 2009) as well as potential host rocks for high-level radioactive waste repositories in several industrial countries (e.g.; Andra, 2005). From their crystalline structure, these clay minerals or phyllosilicates and their corresponding aggregates are thin and aligned “platelets” (Sayers, 1994) that exhibit considerably anisotropic elastic properties which in turn explain the transverse isotropy typical of shales. Thus, the type and amount of clay minerals as well as the geometrical properties of the associated aggregates (shape and mostly orientation) govern the elastic properties and mechanical anisotropy (transverse isotropy, TI) of shales observed in laboratory and the field (Sayers, 1994; Hornby et al., 1994; Hornby, 1998; Sarout and Guéguen, 2008; Vasin et al., 2013). Knowledge of and the ability to predict this TI elastic behavior are critical for seismic prospecting (e.g., Mavko et al., 2009) and for studying the mechanical response of storage in clayey formations (e.g., Giraud et al., 2007).

Nevertheless, shales are rarely pure clayey materials. In addition to interstitial water, shales contain a significant amount of accessory minerals (up to 50 wt%), such as quartz and carbonates, which affect their mechanical properties (Klinkenberg et al., 2009); the content of carbonates and their spatial organization directly impact the failure strength and elastic behavior of shales. An understanding of the elastic behavior of shale requires knowledge of the volume fraction, orientation and connection of the different phases, i.e., the microstructure.

To our knowledge, the effect of microstructure on the elastic properties of shale has been theoretically investigated using two groups of approaches. The first group uses advanced X-ray diffraction and imaging techniques (e.g., scanning electron microscopy-SEM, electronic microprobe analysis, X-ray microtomography and analysis of diffraction images) from which microstructural patterns (grain shapes, pore structure, tortuosity and orientation patterns) are
quantitatively estimated to model the bulk elastic properties. Hornby et al. (1994) and Sayers (1994) pioneered the quantitative estimation of the distribution of clay platelet orientations from digital SEM photographs to model the effective elastic parameters of shale. For this purpose, Hornby et al. (1994) used a sophisticated effective medium approach combining self-consistent (SC) and differential effective medium (DEM) approximations in which the measured orientation distribution functions were used as input parameters. Following this approach, shales were considered as TI porous solids comprising (a) clay-fluid composites oriented at different angles and (b) accessory minerals considered as isolated spherical inclusions randomly dispersed in the mixture, i.e., in the porous solid itself. In an extension of these works, Wenk et al. (2008), and Robinet et al. (2007, 2012) characterized the anisotropy and preferential orientation of minerals of several shales with neutron or X-ray diffraction. The investigated shales exhibited a significant alignment of clay minerals and calcite parallel to the bedding when measuring the orientation distributions from diffraction data: these patterns confirmed the TI behaviour of clay rocks. The authors also extracted quantitative information about phase fractions and their corresponding preferential orientation (i.e., orientation distribution) that were used to model effective properties. However in contrast to Hornby et al. (1994), Wenk et al. (2008) calculated the effective elastic properties by averaging the single-crystal elastic properties over the estimated orientation distribution with simple schemes (i.e., the Voigt and Reuss schemes and the geometric mean). More recently, Vasin et al. (2013) modeled the elastic properties of Kimmeridge shale measured by Hornby (1998) with SC and DEM schemes that used the following microstructural input parameters: (a) mineral volume fractions and orientation distributions obtained via synchrotron X-ray diffraction, (b) pore structure obtained via X-ray tomography and (c) microstructure and nanostructure, of both the minerals and pores obtained via transmission electron microscopy. However, despite the amount of microstructural data obtained and processed, these approaches do not account for spatial correlation between
grains: the accessory minerals (quartz and carbonates) were assumed to be randomly dispersed in the clay phase which was considered as a matrix.

The second group of approaches (Levin and Markov, 2005; Ulm et al., 2005; Giraud et al., 2007) assumes that the quantitative relationships between the microstructure and elastic properties of shale can be captured by identifying the relevant microstructural levels for which the effective properties must be calculated. The objective of this approach is typically to formulate a predictive engineering model. These authors all agree that the following relevant microstructural levels must be considered (Figure 1):

- Level 0: the scale of elementary clay layers is that of mineralogists.
- Level 1 is the scale for which the elementary clay layers are packed together to form a clay particle or aggregate.
- Level 2 is the sub-micrometer scale, often called the “microscopic” scale of porous clay composites constituted in a mixture of clay particles or aggregates. Some authors consider this mixture as a nanogranular material (e.g., Ulm et al., 2005).
- Level 3 often called the “macroscopic” scale is the scale of the characteristic size in the sub-millimeter and millimeter range. At this scale, the rock is viewed as a porous clay matrix mixed with an abundant population of non-clayey grains (mainly quartz, carbonates and pyrite). Numerous theoretical studies have focused on calculating the effective properties associated with this scale (e.g., Hornby et al., 1994).

Following this multi-scale vision, the effective properties at a given level were estimated from the effective properties obtained at lower levels. Different effective medium approximations (EMAs) were used to formulate the effective properties at a given
microstructural level: an SC scheme (Ulm et al., 2005) or Mori-Tanaka (MT) scheme (Giraud et al., 2007).

As for the previous approaches, these multi-scale models do not incorporate statistical and microstructural indicators that account for the relative spatial organization of the clay phase and accessory minerals.

Thus, the main objective of this paper is to quantitatively relate the statistical characterization of the spatial clay distribution and macroscopic elastic properties of shales. This quantitative relationship is achieved by revisiting the Local Porosity Theory (LPT) (Hilfer, 1991;1996) which offers two advantages: (1) it introduces geometric observables such as the clay fraction and clay connectivity which can be easily measured from mineral maps or images and (2) it can be conveniently used for effective medium calculations of elastic properties, as explained below. A local clay theory (LCT), an extension of LPT, is presented in the first part of the paper. The second part illustrates this LCT by numerical applications and a sensitive analysis performed with a real mineral map obtained from Callovo-Oxfordian (COx) mudstone extensively studied for potential repository site of radioactive waste in France.

2. Local clay theory

2.1 Geometric characterization of shales

The starting point of our approach is to consider a shale as a two-phase medium constituted by a clay phase and non-clay phase (i.e., accessory minerals). In terms of the multi-scale structure depicted in Figure 1, the microstructural level considered here is level 3 for which shale is viewed as a porous composite composed of clay and non-clay inclusions. Moreover, the effect of the largest macropores (whose size may be compared to that of accessory minerals) will be omitted: all pores implied in the elastic strain mechanisms of shale are included in the clay phase. This assumption is also physically reasonable because shales contain relatively few
macropores (typically less than 0.5% of the total volume; see e.g., Robinet et al., 2012): their
contribution to the macroscopic elastic properties will also be neglected in a first order
approach.

Following LPT, we do not attempt to identify grains, clay aggregates or other
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another and the same set (they all have the same shape). The local clay fraction \( \phi_c(K_j) \) inside a
measurement cell \( K_j \) can be defined as:

\[
\phi_c(K_j) = \frac{V(C \cap K_j)}{V(K_j)} = \frac{1}{M_j} \sum_{r_i \in K_j} 1_c(r_i)
\]

(1)

where \( V(K_j) \) is the volume of a subset, \( M_j \) denotes the number of volume elements
(voxels or pixels) in \( K_j \) and \( 1_c \) is the characteristic function (indicator function) of clay space
\( C \):

\[
1_c(r_i) = \begin{cases} 
1 & \text{if } r_i \in C \\
0 & \text{otherwise} 
\end{cases}
\]

(2)

From this definition of the local clay fraction \( \phi_c(K_j) \), a simple histogram called local
clay fraction distribution, \( \mu(\phi_c, K) \), can be introduced as follows:

\[
\mu(\phi_c, K) = \frac{1}{M \Delta \phi_c} \sum_{j=1}^{k} \sum_{i=1}^{M_j} (I_{I_i}(\phi_c)I_{I_i}(\phi_c(K_j)))
\]

(3)

or

\[
\mu(\phi_c, K) = \frac{1}{M \Delta \phi_c} \sum_{j=1}^{k} \delta(\phi_c - \phi_c(K_j))
\]

(4)

where \( k \) is the number of classes of histogram, \( I_1, \ldots, I_k \) are the classes of histogram,
\( \Delta \phi_c \) defines the interval width of each class (all classes have the same width), \( 1_{I_i} \) is the indicator
function:

\[
1_{I_i}(\phi_c) = \begin{cases} 
1 & \text{if } \phi_c \in I_i \\
0 & \text{otherwise} 
\end{cases}
\]

(5)

From equation (3), the \( \delta(x) \) function in (4) is defined as follows:

\[
\delta(\phi_c - \phi_c(K_j)) = \sum_{i=1}^{k} (I_{I_i}(\phi_c)I_{I_i}(\phi_c(K_j)))
\]

(6)
In the practical case of a cubical measurement cell $K_j=K(r_j, L)$ of side-length $L$ centred at the lattice vector $r_j$ (i.e., typically a Bravais lattice), the local clay fraction distribution can be rewritten as follows:

$$\mu(\varphi_c, L) = \frac{1}{M \Delta \varphi_c} \sum_{j=1}^{M} \delta(\varphi_c - \varphi_c(r_j, L))$$  \hspace{1cm} (7)

The local clay fraction distribution, $\mu(\varphi_c, L)$, has the following physical meaning: it measures the probability of finding the local clay fraction $\varphi_c$ between $\varphi_c$ and $\varphi_c + d\varphi_c$ in a measurement cell of linear dimension $L$. This distribution allows the mean clay content $\bar{\varphi}_c(L)$ to be defined (if $\Delta \varphi_c \rightarrow d\varphi_c$ with $d\varphi_c << 1$):

$$\bar{\varphi}_c(L) = \int_{0}^{1} \varphi_c \mu(\varphi_c, L) \, d\varphi_c$$  \hspace{1cm} (8)

For an homogeneous shale, the mean clay content $\bar{\varphi}_c(L)$ is independent of $L$ and:

$$\bar{\varphi}_c(L) = \varphi_c(S) = \frac{V(C)}{V(S)}$$  \hspace{1cm} (9)

where the bulk (volumetric) clay content, $\varphi_c(S)$, is the ratio of the volume of the clay fraction, $V(C)$, to the volume of the total sample $V(S)$.

The second geometrical property to characterize the local geometry of shales is related to the local connectivity properties of the clay fraction. Let $\lambda(\varphi_c, L)$ denotes the fraction of percolating measurements cells of side-length $L$ with local clay fraction $\varphi_c$. The local function $\lambda(\varphi_c, L)$ will be called the “local percolation probability” henceforth and is defined as follows:

$$\lambda(\varphi_c, L) = \frac{\sum_{j=1}^{M} \Lambda(r_j, L) \delta(\varphi_c - \varphi_c(r_j, L))}{\sum_{j=1}^{M} \delta(\varphi_c - \varphi_c(r_j, L))}$$  \hspace{1cm} (10)

where the indicator function $\Lambda(r_j, L)$ for the percolation of cell $K(r_j, L)$ is given by:


\begin{equation}
\Lambda(r_j, L) = \begin{cases}
1 & \text{if } K(r_j, L) \text{ percolates in all three directions} \\
0 & \text{otherwise}
\end{cases} \quad (11)
\end{equation}

A measurement cell $K(r_j, L)$ percolates in the $x$- ($y$-, $z$-) direction if there exists a path inside the clay phase connecting two faces of the measurement cell that are perpendicular to the $x$- ($y$-, $z$-) axis. In practice, the function $A(r_j, L)$ can be calculated using the Hoshen-Kopelman algorithm (Hoshen and Kopelman, 1976).

From the particular indicator $A_{\alpha}(r_j, L)$ defined by

\begin{equation}
A_{\alpha}(r_j, L) = \begin{cases}
1 & \text{if } K(r_j, L) \text{ percolates in a direction} \\
0 & \text{otherwise}
\end{cases} \quad (12)
\end{equation}

the $\alpha$-direction percolation probability $\lambda_{\alpha}(\phi_c, L)$ can be calculated as follows:

\begin{equation}
\lambda_{\alpha}(\phi_c, L) = \frac{\sum_{j=1}^{M} \Lambda_{\alpha}(r_j, L) \delta(\phi_c - \phi_c(r_j, L))}{\sum_{j=1}^{M} \delta(\phi_c - \phi_c(r_j, L))} \quad (13)
\end{equation}

From Eq. (10), the total fraction of percolating cells of size $L$ is obtained by integrating over all local clay fractions:

\begin{equation}
p(L) = \int_{0}^{1} \mu(\phi_c, L) \lambda(\phi_c, L) d\phi_c \quad (14)
\end{equation}

The function $p(L)$ characterizes the overall connectivity of the sample at length scale $L$ and is critical for modeling effective properties.

In summary, the two local functions $\mu(\phi_c, L)$ and $\lambda(\phi_c, L)$ are assumed to constitute an approximate but reasonable geometric characterization of the clay distribution in shales (i.e., the local simplicity assumption). By definition, these functions can be calculated from photographs or numerical images of 2D thin sections in a fairly straightforward manner.

However, the local functions $\mu(\phi_c, L)$ and $\lambda(\phi_c, L)$ depend a priori on $L$. As noted by Hilfer (1991), two competing effects should exist. At small $L$ (i.e., $L$ close to the elementary...
voxel/pixel size of the image), the local geometries of measurement cells are highly correlated to each other: the local functions $\mu(\phi_c, L)$ and $\lambda(\phi_c, L)$ cannot capture these complex correlations. At large $L$ (i.e., $L$ close to the size of the shale sample $S$), the local geometries are statistically uncorrelated but each is nearly as complex as the geometry of the full clay distribution of sample $S$. This complexity cannot be described by the local clay function $\phi_c$ and the indicator function for the connectivity $A(r_p, L)$ (i.e., the local simplicity assumption fails).

Moreover, at large $L$, in a practical sense, the number of measurement cells may be low affecting the statistical representativeness of functions $\mu(\phi_c, L)$ and $\lambda(\phi_c, L)$. Thus, we seek an intermediate and optimal scale, termed the critical length $L^*$, at which, the local geometries are uncorrelated and have sufficiently nontrivial geometric content to reasonably capture the spatial fluctuations in the clay content and connectivity. It should be noted that the existence of $L^*$ implies that the shale is assumed to be spatially and statistically homogeneous; the local functions $\mu(\phi_c, L)$ and $\lambda(\phi_c, L)$ do not depend on the position of the measurement cell.

There exist several candidates for the optimal size $L^*$ (Hilfer, 1996; Widjajakusuma et al., 2003): the largest grain size, the length scale at the percolation threshold, the two-point correlation length obtained from the clay fraction auto-correlation function and the percolation length $L_p$ defined from $p(L)$ as follows:

$$\left. \frac{d^2 p}{dL^2} \right|_{L=L_p} = 0$$

Following LPT and Widjajakusuma et al. (2003), the percolation length $L_p$ is the length around which $p(L)$, which is often sigmoidal in shape, changes rapidly from a low value at small $L$ to its trivial value $p(L \to \infty) = 1$ (if clay space $C$ is clearly connected at the scale of $S$). In other words, Eq. (15) defines the domain of the transition between local connectivity (at small $L$) and a global connectivity (at large $L$). Widjajakusuma et al. (2003) demonstrated that LPT provides a reasonable estimate of the effective permittivity at length $L_p$. 
In the following, the optimal size $L^*$ will be considered equal to the percolation length $L_p$ defined by Eq. (15). As noted by Widjajakusuma et al. (2003), this choice offers two advantages: (a) it has been validated for a physical property i.e., permittivity and (b) the calculation of $L_p$ is straightforward.

2.2 Effective medium calculations of elastic properties

The main objective of this section is to demonstrate how the local functions $\mu(\phi_c,L^*)$ and $\lambda(\phi_c,L^*)$ can be connected to conventional EMAs (Figure 2).

For this purpose, following our modeling approach (i.e., LCT), consider our previous partition of a shale sample $S$: a square lattice of $M$ measurements cells of length $L^*$ for which all measurements cells are statistically independent. To build histograms of local functions $\mu(\phi_c,L^*)$ and $\lambda(\phi_c,L^*)$, $k$ classes of local clay fraction $\phi_c^j$ ($\phi_c^j=1, .., \phi_c^{j-k}$) with equal interval $\Delta\phi_c$ must be introduced. For each class of value $\phi_c^j$, the fractions of percolating (measurement) and non-percolating (measurement) cells are $\lambda(\phi_c^j,L^*)\mu(\phi_c^j,L^*)\Delta\phi_c$ and $(1-\lambda(\phi_c^j,L^*))\mu(\phi_c^j,L^*)\Delta\phi_c$, respectively. For the same particular value of $\phi_c^j$, the effective stiffness tensor of the percolating fraction and that of the non-percolating (or blocking) fraction are denoted by $C_p(\phi_c^j)$ and $C_B(\phi_c^j)$, respectively. Bold capital letters refer to fourth-order tensors in the following. As all measurement cells of length $L^*$ are statistically independent, our shale sample $S$ can be considered as a mixture of $2k$ percolating and non-percolating phases associated with the previous $k$ classes of local clay fractions ($\phi_c^{j-1}, .., \phi_c^{j-k}$). The conventional EMAs may then be used for this $2k$-phase mixture.

The first step associated with EMAs is to assume that volume $V$ (surface $S$ for 2D sample) is a representative elementary volume (or a representative elementary surface for a 2D sample); the mechanical response associated with $S$ is assumed to be the macroscopic response
of the shale measured at the laboratory scale. In a second step, in the framework of an elastic behavior, the linear relationship between macroscopic and microscopic strain fields is given as follows:

\[ \varepsilon(r) = A(r) : E \]  

(16)

where \( \varepsilon(r) \) is the microscopic strain tensor; \( E \) is the macroscopic strain tensor i.e., the uniform strain on the boundary of \( V \) and \( A(r) \) is the so-called localization tensor that satisfies:

\[ < A(r) > = I \]  

(17)

with the volume average operator \( < > \) defined, for a physical field, \( l \), by

\[ < l > = \frac{1}{V} \int_{V} l(r) dr \]  

(18)

The tensor \( I \) is the fourth-order symmetric identity tensor. In the framework of elasticity, the effective stiffness tensor \( C^{\text{eff}} \) is introduced as

\[ \Sigma = C^{\text{eff}} : E \]  

(19)

where the macroscopic stress tensor \( \Sigma \) is related the microscopic stress tensor \( \sigma \) by:

\[ \Sigma = < \sigma > \]  

(20)

Obviously, at the microscopic scale:

\[ \sigma(r) = C(r) : \varepsilon(r) \]  

(21)

where \( C(r) \) is the local stiffness tensor. Moreover, according to Eq. (19) and Eq. (21), the effective stiffness tensor \( C^{\text{eff}} \) obeys to the following property:

\[ C^{\text{eff}} : E = \Sigma = < \sigma > = < C : \varepsilon > \]  

(22)

From condition (16), Eq. (22) is rewritten as follows:
According to Eq. (23), the effective stiffness tensor $C^{\text{eff}}$ can be seen as the weighted average of the local stiffness tensor $C(r)$ where the localization tensor $A(r)$ serves as a weighting function. When a mixture of $N$ discrete (perfectly bonded) phases with a volume fraction $f_r$ is considered, Eq. (23) can be expressed as:

$$C^{\text{eff}} = \langle C : A \rangle = \sum_{r=0}^{N} f_r C_r : A_r$$  \hspace{1cm} (24)$$

where $C_r$ is the stiffness tensor of phase $r$ and $A_r$ is the average of the strain localization tensor operator over phase $r$. In our case, the $2k$ volume fractions of percolating and blocking cells correspond to $N$ phases. These $2k$ volume fractions of percolating and blocking cells are then associated with localization tensors of the percolating and blocking cells named $A^j_p$ and $A^j_B$ respectively. Thus, in our case, Eq. (24) is rewritten as follows:

$$C^{\text{eff}} = \sum_{j=0}^{k} \left[ \lambda (\phi^j_c, L^*) \mu(\phi^j_c, L^*) \Delta \phi_c C^j_p : A^j_p + (1 - \lambda (\phi^j_c, L^*)) \mu(\phi^j_c, L^*) \Delta \phi_c C^j_B : A^j_B \right]$$  \hspace{1cm} (25)$$

with the following notations:

$$C^j_p : A^j_p = C_p (\phi^j_c, L^*) : A_p (\phi^j_c, L^*)$$  \hspace{1cm} (26)$$

and

$$C^j_B : A^j_B = C_B (\phi^j_c, L^*) : A_B (\phi^j_c, L^*)$$  \hspace{1cm} (27)$$

The EMA or homogenization scheme used to calculate the effective properties of the percolating and blocking cells, $C^j_p$ and $C^j_B$ are given below (section 2.3).

The localization tensors of the percolating and blocking cells, $A^j_p$ and $A^j_B$ respectively, also depend on the EMA considered to estimate the entire effective property $C^{\text{eff}}$. In the
framework of LPT, Hilfer (1996) suggests to use a SC approach when considering our previous
partitioning \( K \) of the sample using a simple cubic lattice. This choice leads to the following:

\[
\mathbf{C}^{\text{eff}} = \sum_{j=1}^{k} \left[ \lambda(\varphi_c^j, L^*) \mu(\varphi_c^j, L^*) \Delta \varphi_c C_p^j : A_p^{\text{eff,j}} + (1 - \lambda(\varphi_c^j, L^*)) \mu(\varphi_c^j, L^*) \Delta \varphi_c C_c^j : A_c^{\text{eff,j}} \right] \quad (28)
\]

with

\[
A_p^{\text{eff,j}} = (1 + P_p^{\text{eff}} : (C_p^j - C_p^{\text{eff}}))^{-1} \quad (29)
\]

and

\[
A_c^{\text{eff,j}} = (1 + P_c^{\text{eff}} : (C_c^j - C_c^{\text{eff}}))^{-1} \quad (30)
\]

where \( P_p^{\text{eff}} \) and \( P_c^{\text{eff}} \) are the Hill tensors associated with the corresponding SC scheme.

\( P_p^{\text{eff}} \) and \( P_c^{\text{eff}} \) depend on \( C^{\text{eff}} \). Thus, the SC scheme which is inherently implicit requires an
iterative algorithm to obtain \( C^{\text{eff}} \).

A simpler alternative to express \( C^{\text{eff}} \) is likely the Mori-Tanaka (MT) approach which
supposes the existence of an infinite matrix in which all cells are embedded (e.g., Nemat-Nasser
and Hori, 1999). By assuming the presence of a dominant cell phase, the corresponding
effective property \( C^{\text{eff}} \) is now given by (e.g., Nemat-Nasser and Hori, 1999):

\[
\mathbf{C}^{\text{eff}} = \left\{ \sum_{j=1}^{k} \left[ \lambda^j \mu^j \Delta \varphi_c C_p^j : A_p^j + (1 - \lambda^j) \mu^j \Delta \varphi_c C_c^j : A_c^j \right] \right\}^{-1} \left\{ \sum_{j=1}^{k} \left[ \lambda^j \mu^j \Delta \varphi_c A_p^j + (1 - \lambda^j) \mu^j \Delta \varphi_c A_c^j \right] \right\}^{-1} \quad (31)
\]

with the following notations:

\[
\mu^j = \mu(\varphi_c^j, L^*) \quad (32)
\]

\[
\lambda^j = \lambda(\varphi_c^j, L^*) \quad (33)
\]

\[
A_p^j = (1 + P_M : (C_p^j - C_M))^{-1} \quad (34)
\]

\[
A_c^j = (1 + P_M : (C_c^j - C_M))^{-1} \quad (35)
\]
where tensors $C_M$ and $P_M$ are the stiffness tensor of the dominant phase, referred to as the matrix, and the Hill tensor of the dominant phase, respectively. The dominant phase is simply defined as:

$$\mu(\varphi_\epsilon^M, L^*) = \sup_{\varphi' \epsilon \varphi \cdots \varphi I} \left\{ \mu(\varphi_\epsilon^I, L^*) \right\}$$ (36)

Then, the stiffness tensors $C_M$ of the matrix, which is typically percolating, in shales, is:

$$C_M = C_p(\varphi_\epsilon^M, L^*)$$ (37)

The expressions of the components of Hill tensor, $P_M$ are calculated from those of $C_M$ by considering a TI material containing spherical inclusions (e.g., Levin and Markov, 2005; see appendix A).

If $\Delta \phi_k$ tends to zero ($\Delta \phi_k << 1$) (increasing the number $k$ accordingly), the following Riemann integral formulations of Eqs. (28) and (31) are obtained:

$$C^{\text{eff}} = \int_0^1 \left\{ \varepsilon(\varphi_\epsilon, L^*) \mu(\varphi_\epsilon, L^*) C_p(\varphi_\epsilon) : \mathbf{A}_{\epsilon}^{\text{eff}}(\varphi_\epsilon) + (1 - \lambda(\varphi_\epsilon, L^*)) \mu(\varphi_\epsilon, L^*) C_B(\varphi_\epsilon) : \mathbf{A}_{\epsilon B}^{\text{eff}}(\varphi_\epsilon) \right\} d\varphi_\epsilon$$ (38)

(SC approximation)

and

$$C^{\text{eff}} = \int_0^1 \left\{ \varepsilon(\varphi_\epsilon, L^*) \mu(\varphi_\epsilon, L^*) C_p(\varphi_\epsilon) : \mathbf{A}_{\epsilon}^{\text{eff}}(\varphi_\epsilon) + (1 - \lambda(\varphi_\epsilon, L^*)) \mu(\varphi_\epsilon, L^*) C_B(\varphi_\epsilon) : \mathbf{A}_{\epsilon B}^{\text{eff}}(\varphi_\epsilon) \right\} d\varphi_\epsilon \right\}^{-1}$$ (39)

(MT approximation)

Eqs. (38) and (39) involve analytic inversions and multiplications of fourth rank tensors exhibiting TI elastic symmetry. These operations are conveniently performed in terms of a special tensorial basis i.e., $T$-basis (e.g., Levin and Markov, 2005; see Appendix B).
2.3 Effective properties of conductive cells and blocking cells

The mathematical formulations of the stiffness tensors of percolating measurement cells, $C_p(\phi_c, L^*)$ and blocking measurement cells $C_B(\phi_c, L^*)$ (see Eqs. 38 and 39) must be addressed explicitly to use EMAs defined above (Figure 2).

In our case, the stiffness tensor $C_p(\phi_c, L^*)$ associated with percolating measurement cells, is modelled by the following inclusion-based model (Figure 2): a mixture of isotropic spheres embedded in a TI clay matrix. Its counterpart, $C_B(\phi_c, L^*)$, is obtained by considering a mixture of TI and clay spheres embedded in an isotropic mixture constituted in accessory minerals (quartz and calcite). In both cases, the anisotropy is controlled by that of the clay matrix itself.

The effective properties of both sets of mixtures are calculated by a DEM approach, another conventional EMA. The DEM approach offers two advantages. First, this scheme integrates the mechanical interactions between phases in a more realistic way than others EMAs: in particular, the dilute approximation is restricted to low concentration of inclusions whereas the MT approach is limited to moderate concentrations. Second, the DEM approach has been widely and successfully used in rocks physics in order to model the elastic wave velocities of numerous rocks (e.g., Hornby et al., 1994; Markov et al., 2005).

Let us assume that the stiffness tensors, $C_I$ and $C_M$, correspond to the spherical inclusions and matrix of a linear elastic composite medium, respectively. The DEM scheme consists of iteratively adding spherical inclusions with dilute concentration into the current effective medium determined in the previous step. The DEM scheme is relevant for a multiscale structure of inclusional phases; each iteration corresponds to a given inclusion size with a scale separation from the other iterations.
At the initial stage, a material with elastic moduli $C_M$ is considered as the initial host material. A dilute concentration of spherical inclusions is then added into this initial host; the effective tensor of this new composite can be calculated without considering the interactions between inclusions. The construction process continues such that (a) at each stage, the embedded spherical inclusions are in dilute concentration and (b) the required volume fraction of inclusions is satisfied. At a given step indexed $i$, the small increment of stiffness due to the addition of a small fraction of inclusions, $df_i$ is given by (e.g., Nemat-Nasser and Hori, 1999):

$$
\frac{dC_{i}^{\text{eff}}}{I - f_i} (C_I - C_{i}^{\text{eff}}) \left[ I + P_i : (C_I - C_{i}^{\text{eff}}) \right]
$$

(40)

This increment $dC_{i}^{\text{eff}}$ must be added to the effective tensor $C_{i}^{\text{eff}}$ obtained in the previous step. The Hill’s tensor is a function of $C_{i}^{\text{eff}}$ and the Eshelby’s tensor which depends on the chosen inclusion-based model:

$$
P_i = S_i : (C_{i}^{\text{eff}})^{-1}
$$

(41)

When percolating measurement cells are considered, the stiffness tensors $C_I$ and $C_M$, are associated with isotropic spheres and a TI-clay matrix, respectively. When the effective tensors of blocking measurement cells are calculated, the tensor $C_I$ is that of TI-clay spheres embedded in an isotropic matrix of stiffness $C_M$ (Figure 2).

3. Numerical results

3.1 Numerical application to Callovo-Oxfordian mudstone

3.1.1. Sample description and mineral map

The LCT presented in the previous section and summarized in Figure 2 is now applied to a mineral map made from a Callovo-Oxfordian mudstone sample.
The Callovo-Oxfordian mudstone involved in this study is a clayey formation at the site of the French Meuse/Haute-Marne Underground Research Laboratory (MHM-URL). The thickness of this formation is 130 m and its age 150-160 My, the formation is located 420-550 m below the surface, in the eastern part of the Paris Basin (Andra, 2005). The Callovo-Oxfordian formation contains mainly 25 to 65 wt.% clay minerals, with 20-42 wt.% carbonates (calcite, dolomite, ankerite) and 15-31 wt.% quartz and feldspars (Andra, 2005).

The mineral map used in this study was prepared from a drill-core, denoted as EST05-709 (-492.2 m) and extracted from the Andra EST205 borehole. The rough mineral composition estimated by XRD and the petrophysical properties measured in this sample are given in Table 1. The velocity measurements were carried using an ultrasonic (1MHz) pulse transmission technique to obtain P and perpendicularly polarized shear wave velocities (e.g., Jorand, 2006, Ghorbani et al., 2009).

The mineral map was obtained at a micrometer spatial resolution from an advanced image processing of a chemical elements map that was acquired through the use of a Cameca SX100 electron probe microanalyzer (Prêt et al., 2010). This electron microanalyze provides quantitative concentration maps of 14 chemical elements (Al, Na, K, Ca, Si, Mg, Ti, Fe, S, Ba, Zr, P, Zn, Sr) on a 3 x 0.5 mm² area with a spatial resolution of 2 μm/pixel. The image processing of these maps is based on mineral thresholding methods that accommodate mixtures and solid solutions. For details, the reader is referred to Prêt et al. (2010). In our case, this methodology allows for the spatial location of 16 different minerals, including three different clay minerals: illite-smectite mixed layers, kaolinite and chlorite. No pure illite was detected but two micas were recognized: glauconite and muscovite. The result of this image analysis is given in a numerical file in which the location and mineral code of each pixel of the mineral map are indicated. This numerical file constitutes the input file for calculations of local geometry distributions associated with LCT.
A region of interest and the properties of the corresponding map are given in Figures 3 and Table 2, respectively. This map was prepared from a polished thin section in a plane perpendicular to the stratigraphic plane. The x and y- directions indicated in Figures 3 are parallel to the bedding planes whereas the z-direction is perpendicular to the bedding.

The content of a given mineral indicated in Table 2 is the ratio of the number of pixels of the given mineral divided by the total number of pixels. Consequently, Table 2 indicates mineral composition given in terms of “surface” contents (equal to volumetric contents for a TI material observed in a plane perpendicular to the stratigraphic plane) whereas mineral contents are expressed in weight per-cent from an estimation provided by XRD (Table 1), which explains the differences observed between Tables 1 and 2.

3.1.2 Calculations of the local geometry distributions

The mineral map is now used to calculate the local functions associated with LCT, namely the local clay fraction distribution, \( \mu(\phi, L) \), and local percolation probability, \( \lambda(\phi, L) \).

Figure 4 illustrates these calculations for different sizes of measurement cell \( L \). For clarity, the histograms were calculated with a large interval \( \Delta \phi = 0.02 \) and smoothed with a Stine man function. These local functions which depend significantly on \( L \) exhibit two features.

First, the local clay fraction distributions \( \mu(\phi, L) \) calculated for \( L \) greater than 10 pixels appear roughly as Gaussian-type curves centred on the mean “surface” clay content \( \bar{\phi}_c \) (i.e., \( \text{Jora} \bar{\phi}_c = 50.44\% \), see Table 2). This mean or “peak” value which corresponds to a maximum value in the local clay fraction distributions can be considered as a dominant phase or a matrix when all of the measurement cells are percolating (i.e., \( \lambda(\phi_c, L) = 1 \) ): in other words, this dominant phase has a specific morphological matrix role. This observation will further justify the MT scheme which assumes the existence of a matrix. Moreover, in a second-order analysis,
these local clay fraction distributions exhibit slight dissymmetry towards high values of $\varphi_c$. A similar dissymmetry was also observed in the local porosity distributions obtained from sedimentary formations in the framework of the LPT (Widjajakusuma et al., 1999).

Second, the calculated local percolation probabilities $\lambda(\varphi_c,L)$ and corresponding connectivities increase with $L$ (Figure 5). Moreover, they indicate that a minority fraction of measurement cells at clay fraction $\varphi_c = \bar{\varphi}_c$ percolates when $L$ is small (e.g., 10 pixels). For larger $L$ values of approximately 50 pixels, almost half of the measurement cells percolate at the mean-peak value $\bar{\varphi}_c$ . In other words, all the measurement cells percolate (i.e., $\lambda(\varphi_c,L) = 1$ ) for values of $\varphi_c$ that are considerably higher than the mean-peak value $\bar{\varphi}_c$ calculated from the mineral map.

To calculate the effective property of this mineral map, the total fractions of percolating cells $p(L)$ and the corresponding critical length $L^*$ must be determined. Three domains can be identified from the results given in Figure 6. The first domain (Domain 1) is associated with a “local” connectivity (length-sizes less than 40 pixels i.e., 80 $\mu$m). In this domain, the total fraction of percolating cells increases significantly with $L$. The domain of large $L$ (Domain 3) is related to a “global” connectivity of measurement cells whose length-sizes, greater than 75 pixels (i.e., 150 $\mu$m), are close to that of the mineral map. Between these domains, the intermediate domain 2 exhibits a plateau for which the criterion defined by Eq. (15) can be satisfied. As a result and based on the criterion in Eq. (15), a critical length $L^*$ of $55 \pm 5$ pixels ($110 \pm 10$ $\mu$m) is obtained (Figure 6).

### 3.1.3 Calculations of the effective elastic properties

A value of 55 for $L^*$ is now used to model the effective elastic properties of the shale sample. Two groups of input data are required for this purpose: (a) local functions associated
with LCT, namely $\mu(\phi_c, L^*)$ and $\lambda(\phi_c, L^*)$ both deduced from the mineral map, and (b) the elastic properties of the two phases constituting the shale sample: those of the clay matrix and those of
the non-clay phase.

For the first group of input data, the local clay fraction distribution, $\mu(\phi_c, L^* = 55)$, is approximated by a Gaussian distribution (Figure 4):

$$\tilde{\mu}(\phi_c) = \frac{1}{\sigma_1 \sqrt{2\pi}} e^{-\frac{1}{2} \left( \frac{\phi_c - \phi_1}{\sigma_1} \right)^2}$$

(42)

with the following best-fitting values: $\phi_1 = \bar{\phi}_c = 0.504$ and $\sigma_1 = 0.052$ ($R^2 = 0.955$) (Figure 7). Parameter $\sigma_1$ often called the standard deviation, controls the width of the Gaussian curve.

To support the gaussianity assessment, a test for normality (e.g., Shapiro-Wilk test) is applied to a set of 13 $\mu(\phi_c, L^*)$ data values centred on the mean “surface” clay content $\bar{\phi}_c$. The result of the Shapiro-Wilk test, performed using Excel, indicates a p value greater than the level of significance of 5% ($\alpha = 0.05$): these data can be considered as normally distributed.

The local percolation probability, $\lambda(\phi_c, L^* = 55)$ is assumed to be a simple step or sigmoid function and is written as follows (Figure 5):

$$\tilde{\lambda}(\phi_c) = \frac{1}{1 + e^{-\frac{\phi_c - \phi_2}{\sigma_2}}}$$

(43)

with best-fitting values of: $\phi_2 = 0.494$ and $\sigma_2 = 0.031$ ($R^2 = 0.959$) (Figure 7).

In the second group of input data, the elastic moduli of non-clay phases (quartz and calcite) are values taken from Mavko et al. (2009). In our calculations, the isotropic elastic moduli of the non-clay phase, i.e., the bulk and shear modulus, are weighted averages of those of quartz and calcite where the weights are the gravimetric contents of each phase as noted in Table 2 (see the results in Table 3). In fact, pure calcite and quartz phases are anisotropic materials: the isotropized equivalent elastic properties considered here correspond to random
distributions in orientations and space of calcite and quartz inclusions. This assumption has been used by many other authors (e.g., Ulm et al., 2005; Giraud et al., 2007).

The elastic moduli of the clay phase are more difficult to estimate, because the clay phase is also a porous and heterogeneous composite composed of aggregates of clay layers, pores filled with interstitial water and likely fine non-clay minerals (Figure 1). Therefore, a range of elastic moduli for the clay phase will be considered in the following. This range is estimated from the previous work by Ulm which used an experimental micromechanics approach, combining nanoindentation measurements with microporoelasticity theory (Ulm et al., 2005). Indeed, both bounds proposed in Table 3 result from a micromechanical inversion of elastic moduli from nanoindentation measurements obtained on three different shale materials. These bounds are inverted values of the elastic moduli of the clay phase considering range of spatial scales of $10^{-7}$-$10^{-5}$ m (typically the microstructural levels 1 and 2 defined in Figure 1) of Ulm’s work; the elementary pixel size of our mineral map is 2 μm. The values in Table 3 indicate a large range of possible values of TI elastic moduli confirming that the elasticity of the clay phase is poorly constrained in our context.

These inverted poroelastic parameters are undrained stiffness values because the calculated effective elastic properties will be compared to dynamic (ultrasonic) measurements.

3.1.4 Comparison with the measured macroscopic data

In this section, some of the effective elastic properties inferred from the mineral map of sample EST05-709 are compared to those measured on the same sample by a macroscopic ultrasonic velocity method (e.g., Jorand, 2006, Ghorbani et al., 2009). More precisely, the measured TI elastic moduli $C_{11}^{mes}$ and $C_{66}^{mes}$ are obtained as follows (e.g.; Sarout and Guéguen, 2008):

$$C_{11}^{mes} = \rho V_{P}^{2}(0^\circ) = \rho V_{PH}^{2}$$  \hspace{1cm} (44)
\[ C_{66}^{mes} = \rho V_S^2(0^\circ) = \rho V_{SH}^2 \]  
(45)

where \( V_S^2(0^\circ) = V_{PH}^2 \) is the bedding-parallel compressional velocity, \( V_S^2(0^\circ) = V_{SH}^2 \) is the horizontally polarized, bedding-parallel shear velocity and \( \rho \) is the rock density. The Voigt notation is here used here (see Appendix A) and thus, elastic modulus \( C_{12}^{mes} \) can be inferred from the previous moduli as follows:

\[ C_{12}^{mes} = C_{11}^{mes} - 2C_{66}^{mes} \]  
(46)

The values of \( C_{11}^{mes} \) and \( C_{12}^{mes} \) calculated from (44) and (46) are given in Table 4. The three other elastic moduli, \( C_{13} \), \( C_{33} \) and \( C_{44} \) have not been calculated because the corresponding experimental velocities have not been measured.

The experimental values of \( C_{11}^{mes} \) and \( C_{12}^{mes} \) indicated in Table 4 are significant lower than the upper bounds of the elastic moduli for the clay phase proposed in Table 3. Thus, the latter cannot be used as input data to model the effective properties of sample EST05-709. For this purpose, the lower bounds in Table 3 are preferred.

These lower bounds of the elastic moduli of the clay phase and the MT approach (Eq. 35 solved by the Romberg’s method implemented in a Fortran90 program) are now used to estimate the effective elastic constants of sample EST05-709. Two values of the dominant clay fractions, \( \phi_c \), are considered: (i) the value of 0.504 which corresponds to the “surface” value calculated from the corresponding mineral map (see Table 2) and (ii) that of 0.569 calculated from the (bulk) gravimetric clay content, \( w_c = 0.65 \) (see Table 1), of sample EST05-709 as follows:

\[ \phi_c = \frac{\rho}{\rho_c} w_c \]  
(47)

where \( \rho \) is the shale density equal to 2.32 g.cm\(^{-3}\) (Table 1) and \( \rho_c \) is the clay density equal to 2.65 g.cm\(^{-3}\).
For both values of the dominant clay fractions, $\bar{\phi}_c$, our results in Table 4 indicate that the effective elastic constants estimated by LCT are on the same order of magnitude that those given by ultrasonic velocity measurements. The absolute relative errors for $C_{11}$ and $C_{12}$ are 12.1 and 32.6%, respectively. Moreover, Table 5 provides the best fit values of the elastic properties of the clay phase, which were used as input data to model our experimental data. These elastic constants were simply inferred by a bounding approach.

The calculations presented in Tables 4 and 5 do not validate the proposed LCT: they simply illustrate the capability of the theoretical framework promoted in this work to model macroscopic and elastic properties. To proceed, as previously and implicitly noted in Table 5, elastic constants of clay phase must be independently and experimentally constrained, i.e., measured accurately.

3.2 Sensitivity analysis of the local geometry distributions

a- Influence of the local clay fraction distribution $\mu(\phi_c; L^*)$

In this section, the impact of spatial clay fluctuation on the effective elastic constants of shale is investigated via LCT. As mentioned previously, following our theoretical framework, the spatial clay fluctuation is considered by the local clay fraction distribution $\mu(\phi_c; L^*)$ which will take the particular form of a Gaussian function (i.e., the normal distribution given by Eq. 42). This choice is justified by our experimental data (see Figure 7) and by simplicity. But we are aware that other distributions, for instance beta function whose support is [0,1] could be used. At this stage, no experimental argument allows to promote a particular distribution.

The amplitude of the spatial clay fluctuation will be captured by the standard deviation $\sigma_1$ of the Gaussian function in Eq. (42): a more important spatial clay fluctuation will have a higher standard deviation $\sigma_1$. In other words, the effective elastic constants of shale will be
calculated as a function of $\sigma_1$ and by considering two values for the mean of the Gaussian distribution: $\bar{\phi}_c^1 = 0.5$ (50%) and $\bar{\phi}_c^2 = 0.6$ (60%).

Moreover, the values of the standard deviation $\sigma_1$ are chosen in such a way that (see Eq. 5):

$$\int_0^1 \phi_c \mu(\phi_c, L^*) \, d\phi_c = \bar{\phi}_c^{1or2}$$ (48)

This condition will ensure that all cases of this sensitivity analysis will exhibit the same mean clay content ($\bar{\phi}_c^1$ or $\bar{\phi}_c^2$); the sole spatial clay fluctuation around the mean value will be studied. For the case in which $\bar{\phi}_c^1 = 0.5$, condition (48) is satisfied if the standard deviation $\sigma_1$ is in the range of $[0, 0.15]$. For the case in which $\bar{\phi}_c^2 = 0.6$, the standard deviation $\sigma_1$ has to be in the range of $[0, 0.13]$ (Figure 8).

The elastic constants of a soft clay matrix, a hard clay matrix (see values in Table 3) and a single non-clay phase (quartz) will be considered in these calculations. The bulk and the shear moduli of quartz are set equal to 37.0 and 44.7 GPa, respectively (Mavko et al., 2009).

This sensitivity analysis is conducted by calculating and displaying a normalized deviation parameter, $\Delta_{ij}$, defined as follows:

$$\Delta_{ij} = \frac{C_{ij}^{LCT} - C_{ij}^{U}}{C_{ij}^{U}}$$ (49)

where $C_{ij}^{LCT}$ are the effective elastic properties calculated by LCT and $C_{ij}^{U}$ are the effective elastic properties calculated in the uniform case (i.e., with no spatial clay fluctuation: $\mu(\phi_c, L^*) = \delta(\phi_c - \overline{\phi}_c)$) and by using the previous DEM approach that consider percolating cells (section 2.3). Indeed, in the calculations performed in this section, all measurement cells are
percolating (i.e., \( \lambda(\phi_c, L^*) = 1 \)). Such an ideal and simple case mimics an ideal dispersion of non-clay grains without accounting for potential clustering. The case for which the clay matrix is partially non-percolating (\( \lambda(\phi_c, L^*) \neq 1 \)) will be investigated in the next section.

In the TI case, following Voigt notation, the normalized deviation parameter, \( \Delta_{ij} \) is calculated for the five independent components of the TI elastic tensors involved in Eq. (49) (i.e., \( \Delta = \{\Delta_{11}, \Delta_{12}, \Delta_{13}, \Delta_{33}, \Delta_{44}\} \)).

Figures 9a,b,c, d and e display the results of this sensitivity analysis for both mean clay contents (\( \bar{\phi}_c^1 \) and \( \bar{\phi}_c^2 \)) considering the elastic constants of a soft and hard clay matrix, respectively (see Table 3). These plots indicate the following two major features:

- The \( \Delta_{ij} \) parameters exhibit different sensitivities to the mean clay content changes and the variation in elastic rigidity of the clay matrix (compare \( \Delta_{11} \) and \( \Delta_{12} \) to \( \Delta_{33} \) and \( \Delta_{44} \)). Moreover in some cases (for instance \( \Delta_{33} \) and \( \Delta_{44} \) when \( \bar{\phi}_c^2 = 60\% \)), the \( \Delta_{ij} \) evolutions are not monotonic functions of \( \sigma_1 \). The evolutions often exhibit a maximum that appears to be a function of the mean clay content and the elastic rigidity of the clay matrix.

- However, the \( \Delta_{ij} \) values are low i.e., less than 2% for all values of mean clay content (\( \bar{\phi}_c^1 \) or \( \bar{\phi}_c^2 \)) and clay matrix rigidity; the spatial clay fluctuation has a small impact on the shale elasticity, when all the cells are percolating.

For the first feature, the results suggest that \( \Delta_{ij} \) parameters depend primarily on the contrast between the clay matrix elasticity and that of the non-clay phase and to a lesser extent on the mean clay fraction \( \bar{\phi}_c \). Indeed, considering parameters \( \Delta_{13}, \Delta_{33} \) and \( \Delta_{44} \), for which the clay matrix constants of the hard and soft clay matrices are similar (see Table 3), the evolutions of these deviation parameters as a function of \( \sigma_1 \) are similar up to a threshold of approximately
13% (Figure 9c, d and e). After this threshold, these evolutions are different and are governed mainly by the mean clay fraction $\bar{\phi}_c$. In contrast for parameters $\Delta_{11}$ and $\Delta_{12}$, for which the clay matrix constants $C_{11}$ and $C_{12}$, of the hard and soft clay matrices are different (see Table 3), the evolutions of these deviation parameters are mainly governed by these contrasts; the differences in the mean clay content have a much smaller impact, and parameters $\Delta_{11}$ and $\Delta_{12}$, are greater when a soft matrix is considered. This last observation also suggests that spatial clay fluctuation have a greater impact on the effective properties when there is a high elastic contrast between clay matrix and non-clay phase.

However, as noted above, this impact is low in magnitude (below 2%). We now explore whether this feature could be explained by a first order approach, from the symmetric nature of the local clay fraction distribution $\mu$. Indeed, the number of cells with clay fractions less than the mean-peak value $\bar{\phi}_c$ is similar to that with clay fractions greater than $\bar{\phi}_c$, and thus, the effects associated with both opposite populations of measurement cells would be compensated; the LCT calculations would lead to effective properties that are highly similar to the average values obtained from the mean value $\bar{\phi}_c$. The following simple asymmetric distribution has been considered to confirm the validity of this statement:

$$
\tilde{\mu}(\phi_c) = \frac{\sqrt{2}}{(\sigma_1^+ + \sigma_1^-)\sqrt{\pi}} \left[ e^{-\frac{1}{2}\left(\frac{\phi_c - \phi_{\text{peak}1}}{\sigma_1}\right)^2} \times 1_{[-\infty, \phi_{\text{peak}}]}(\phi_c) + e^{-\frac{1}{2}\left(\frac{\phi_c - \phi_{\text{peak}1}}{\sigma_1^-}\right)^2} \times 1_{[\phi_{\text{peak}}, +\infty]}(\phi_c) \right]
$$

(50)

where

$$
1_{[-\infty, \phi_{\text{peak}}]}(\phi_c) = \begin{cases} 
1 & \text{if } \phi_c \leq \phi_{\text{peak}} \\
0 & \text{otherwise}
\end{cases} \quad \text{and} \quad 1_{[\phi_{\text{peak}}, +\infty]}(\phi_c) = \begin{cases} 
1 & \text{if } \phi_c > \phi_{\text{peak}} \\
0 & \text{otherwise}
\end{cases}
$$

This asymmetric distribution is a simple extension of the general Gaussian distribution defined by (38). In this sensitivity analysis, parameter $\sigma_1^+$ was kept constant and equal to 0.05.
and the parameter $\sigma^-_1$ was increased accordingly (Figure 10). The $\varphi_{\text{peak}}$ values in (46) were calculated for each value of $\sigma^-_1$ and $\sigma^+_1$ from the following equation which defines the mathematical formulation of the mean clay fraction $\bar{\varphi}_c$ of this asymmetric distribution:

$$
\varphi_{\text{peak}} = \bar{\varphi}_c - \frac{2}{\sqrt{\pi}} (\sigma^+_1 - \sigma^-_1)
$$

(51)

where, the same mean clay fraction $\bar{\varphi}_c$ equal to 60% was considered in all results depicted in Figure 10 for comparison purposes. These results must be compared to the symmetric case (no skewness): $\sigma^+_1 = \sigma^-_1 = 0.05$. As illustrated in Figure 10, a significant increase in most $\Delta_{ij}$ parameters with the skewness of $\mu$ is observed: for instance, the $\Delta_{44}$ parameter increases from 0.3 to 4.5% when $\sigma^-_1$ increases from 0.05 (no skewness) to 0.25. However, all of these $\Delta_{ij}$ values are still low (less than 5%): the symmetric shape of the local clay fraction distribution can explain only a part of the low impact of spatial clay fluctuation on the effective properties, when all measurement cells are percolating.

**b- Influence of the local percolation probability $\lambda(\varphi_c,L^*)$**

In this section, we still assumed that local clay fraction distribution $\mu(\varphi_c,L^*)$ is a normal distribution given by Eq. (42). The influence of spatial connectivity fluctuations is now investigated by using the sigmoid function given in (43). This simple sigmoid function which fits our experimental data (Figure 7), is described by two parameters.

The first parameter, $\varphi_2$, defines the location of the inflexion point and, thus that of the ramp in the $\varphi_c$-axis. If this parameter is considerably higher (lower) than the mean clay fraction, $\bar{\varphi}_c$, then most measurements cells are blocking (percolating) cells (Figure 11a).
The second parameter $\sigma_2$ defines the width of the sigmoid or the S-curve; lower $\sigma_2$ values cause narrow S-curves and the uniformity of spatial connectivity is more important (Figure 11b).

Both parameters are considered in the sensitivity analysis presented below. This sensitivity analysis consists of calculating the normalized deviation parameter, $\Delta_y$, defined in Eq. (49) for various values of $\varphi_2$ and $\sigma_2$; the parameter $\varphi_2$ varies in the range [0.4-0.8] and the second parameter $\sigma_2$ is considered to belong to the range [0.05-0.2]. Two Gaussian local clay fraction distributions, $\mu$, are considered in these numerical simulations: a narrow distribution with a standard deviation $\sigma_1$ of 0.05 and a broad distribution with a standard deviation $\sigma_1$ of 0.15. In all cases, the mean-peak value $\overline{\varphi}_c$ of the Gaussian local clay fraction distributions is held constant and equal to 0.6 (60%). As in the previous section, the clay matrix exhibits two elastic rigidities i.e., a “soft” clay matrix and a “hard” clay matrix are considered (see values of elastic constants in Table 3). The non-clay phase is quartz.

Figures 12a,b,c,d and e display the $\Delta_y$ values for the variation range of parameter $\varphi_2$ considered in this sensitivity analysis. The second parameter $\sigma_2$ was equal to 0.05. The $\Delta_y$ curves displayed in the figures 12a,b,c,d and e indicate three main results.

First, for all values of clay matrix rigidity and standard deviation, $\sigma_1$ (i.e., the width of distribution $\mu$), the normalized deviation parameter, $\Delta_y$, is an increasing and quasi-sigmoidal function of parameter $\varphi_2$; higher $\varphi_2$ values imply greater deviations in the effective constants from the percolating and reference case given by $C^{ij}_{ij}$ (i.e., the inclusion model with a non-clay spherical grain embedded in a TI clay matrix without spatial clay and spatial connectivity fluctuations). This observation is not surprising. When $\varphi_2 \ll \overline{\varphi}_c$, most measurement cells are
percolating (i.e., $\lambda(\varphi_c,L^*)=1$) and the cases described in the previous section 3.2.a are retrieved: the $\Delta_{ij}$ values are low (Figures 10a,b,c,d and e). When $\varphi_2 \gg \varphi_c$, the effective properties deviate from the percolating and reference case, $C_{ij}^{U}$ to tend to the elastic constants of the blocking inclusion-based model (i.e., the inclusion-based model with a TI clay grain embedded in an isotropic non-clay matrix). When $\varphi_2 = \varphi_c$, the $\Delta_{ij}$ values may be rather high reaching values close to 30% (Figure 12e). In other words, even if the clay fraction is dominant ($\varphi_c = 0.6$=60%), a simple percolating inclusion-based model in which a non-clay spherical grain is embedded in a TI clay matrix may lead to significant errors in the prediction of the effective properties of shales.

Second, when $\varphi_2 > \varphi_c$ and for all values of standard deviation $\sigma_1$ (i.e., the width of distribution $\mu$), the $\Delta_{ij}$ values of shale associated with a “soft” clay matrix are greater than those associated with a “hard” clay matrix. In other words, when $\varphi_2 > \varphi_c$ (i.e., most measurement cells are not percolating), high contrast between the elastic rigidities of the clay matrix and grain increase the deviation between the predicted effective properties and those of the percolating and reference case. This statement is confirmed for $\Delta_{11}$ and $\Delta_{12}$ at all $\varphi_2$ values.

Third, for a given elastic rigidity of the clay matrix, the $\Delta_{ij}$ values of shale associated with a broad distribution of $\mu$ (i.e., $\sigma_1 = 0.15$) are greater than those associated with a narrow distribution of $\mu$ (i.e., $\sigma_1 = 0.05$) up to a $\varphi_c$ value close to 0.64, after which the trend is reversed. Thus, when $\varphi_2 = \varphi_c$, for a given elastic rigidity of the clay matrix, a broad distribution of $\mu$ leads to a more significant deviation from the percolating and reference case.

Figures 13a,b,c,d and e display the $\Delta_{ij}$ values for the variation range of parameter $\sigma_2$.

Parameter $\varphi_2$ was kept equal to 0.6 (60%). The results indicate that the normalized deviation
parameter, $\Delta_{ij}$, is not highly sensitive to changes in $\sigma_2$; a small decrease of $\Delta_{ij}$ with $\sigma_2$ is observable for most cases. Thus, the deviation between the calculated effective properties and those of the percolating and reference case depends only slightly on the spreading of clay phase connectivity.

4. Conclusion

In our opinion, this paper is primarily of methodological interest. It introduced a LCT whose theoretical foundations were provided in the Hilfer’s Local Porosity Theory, to establish a quantitative relationship between the effective (macroscopic) elastic properties and the stochastic character of the clay fraction and clay percolation functions of shales. This theoretical approach is based on three assumptions. First, shale can be considered as a two-phase medium composed of clay and non-clay-phases. Second, a local simplicity assumption implies that the complexity of the spatial clay distribution can be captured by two local functions $\mu(\phi_c,L)$ and $\lambda(\phi_c,L)$ calculated from a partitioning of a mineral map ($L$: the map side-length) and a local clay fraction $\phi_c$ of each subset called measurement cell. The local clay fraction distribution $\mu(\phi_c,L)$ provides information about spatial clay fluctuations and the local percolation probability $\lambda(\phi_c,L)$ describes the spatial fluctuations of clay connectivity. These two local functions can only be captured through the use of mapping techniques whereas bulk methods such as XRD only provide a mean clay fraction. Third, there exists a side-length $L^*$ for which the local geometries given by local functions $\mu(\phi_c,L^*)$ and $\lambda(\phi_c,L^*)$ in each measurement cells are uncorrelated with each other. This assumption allows for the connection of both local functions for EMAs (here, a DEM scheme).
This LCT was applied to a sample of the Callovo-Oxfordian (COx) mudstone, for which a mineral map and petrophysical data were available. The area of the mineral map was 3 x 0.5 mm² with a spatial resolution of 2 μm/pixel. Two results were obtained from this application. First, the size and the spatial resolution of the mineral map were adequate to extract the local functions $\mu(\phi_c,L^*)$ and $\lambda(\phi_c,L^*)$ and thus to calculate effective elastic properties. Second, it was demonstrated that the local functions $\mu$ and $\lambda$ could be reasonably approximated by Gaussian and simple sigmoid functions, respectively. Although it does not fully validate the LCT promoted in this work, this application illustrates the promising ability of this theoretical approach to infer macroscopic properties from microstructural data, (in our case, mineral maps).

LCT was also used in a sensitivity analysis to evaluate the potential effects of spatial fluctuations in the clay phase and clay conductivity on the effective elastic properties of shales. These effects were investigated by comparing the effective properties calculated from LCT to those provided by a simple percolating inclusion-based model: a non-clay spherical grain embedded in a TI clay matrix.

If the local function $\mu(\phi_c,L^*)$ is approximated by a Gaussian-type function with standard deviation in the range [0.05-0.19], the results have shown that the spatial clay fluctuations (through $\mu(\phi_c,L^*)$) have only a slight impact on the calculated effective properties; in our simulations, these fluctuations lead to quantitative deviations from the inferred effective properties and those of the percolating inclusion-based model, lower than 5%. However this result is not reproduced when the spatial fluctuations of clay connectivity (through $\lambda(\phi_c,L^*)$) are considered. When a simple sigmoid function is used to describe the local percolation probability $\lambda$, the deviations from the inferred effective properties and those of the percolating inclusion-based model can exceed 50%. Particularly when the sigmoid function is centered on the mean-peak value of the local clay fraction distribution $\mu$, the percolating inclusion-based
model leads to significant errors (up to 30%) in the prediction of the effective properties of shales.

This work strongly suggests the need for further investigations in two directions. First, this work has accounted for simple microstructural patterns but can be extended by incorporating additional microscopic features (e.g., orientation of non-clay grains) which would have been quantified (e.g., Robinet et al., 2012). Thus, this work confirms the interest of microstructural investigations and mapping techniques to identify the microscopic and geometric factors controlling the effective properties measured in the field. Second, our sensitivity analysis has highlighted the crucial importance of the elastic constants of the clay matrix in calculations of effective properties. The direct and accurate measurement of these particular mechanical parameters is still a challenging area of experimental research.

Appendix A. Hill polarization tensor for a spherical inclusion in a transversely isotropic medium

The expressions of components of the Hill’s tensor $P_M$ for a spherical inclusion in a transversely isotropic medium are taken from Withers (1989) and Giraud et al. (2007).

Following the Voigt notation, the elastic constants of the transversely clay matrix are referred as $C_{11}$, $C_{33}$, $C_{12}$, $C_{13}$ and $C_{44}$. Similarly as in Withers (1989), the following parameters are introduced:

$$ C_{13}^* = (C_{11}C_{33})^{1/2}, \quad k_i = \frac{C_{11}/\nu_i^2 - C_{44}}{C_{13} + C_{44}}, $$

$$ D = \frac{1}{4\pi C_{44}\nu_3} \quad (A1) $$

$$ \nu_1 = \left[ \frac{(C_{13}^* - C_{13})(C_{13}^* + C_{13} + 2C_{44})}{4C_{33}C_{44}} \right]^{1/2} + \left[ \frac{(C_{13}^* - C_{13})(C_{13}^* - C_{13} - 2C_{44})}{4C_{33}C_{44}} \right]^{1/2} \quad (A2) $$

34
\[ v_2 = \left( \frac{(C_{13}^* - C_{13})(C_{13}^* + C_{13}^2 + 2C_{44})}{4C_{33}C_{44}} \right)^{1/2} - \left( \frac{(C_{13}^* - C_{13})(C_{13}^* - C_{13}^2 - 2C_{44})}{4C_{33}C_{44}} \right)^{1/2} \]  

(A3)

\[ v_3 = \left( \frac{C_{66}}{C_{44}} \right)^{1/2} \text{ with } C_{66} = \frac{C_{11} - C_{12}}{2} \]  

(A4)

At this stage, two cases have to be considered.

(a) if \( C_{13}^* - C_{13} - 2C_{44} \neq 0 \), the components of the Hill’s tensor \( P_M \) are expressed as follows (Withers, 1989):

\[ P_{11} = \frac{3}{2} \sum_{i=1}^{2} v_i A_i' I_1(i) + \frac{D I_1(3)}{4}, \quad P_{12} = \frac{1}{2} \sum_{i=1}^{2} v_i A_i' I_1(i) - \frac{D I_1(3)}{4} \]  

(A5)

\[ P_{13} = \sum_{i=1}^{2} k_i v_i^3 A_i' I_2(i), \quad P_{33} = -2 \sum_{i=1}^{2} k_i v_i^5 A_i' I_2(i) \]  

(A6)

\[ P_{44} = \frac{1}{4} \sum_{i=1}^{2} (1 + k_i) v_i^3 A_i' \left[ I_2(i) - 2k_i I_1(i) \right] + \frac{D v_i^2 I_2(3)}{8} \]  

(A7)

with

\[ A_1' = \frac{C_{44} - v_1^2 C_{33}}{8\pi v_1^2 v_1^3 C_{33} C_{44}}, \quad A_2' = -\frac{C_{44} - v_2^2 C_{33}}{8\pi v_2^2 v_2^3 C_{33} C_{44}} \]  

(A8)

For the sake of clarity, the subscript \( M \) associated with the components of the Hill’s tensor has been omitted.

(a) if \( C_{13}^* - C_{13} - 2C_{44} = 0 \), the components of the Hill’s tensor \( P_M \) are given by (Giraud et al., 2007):

\[ P_{11} = 3 \left( \frac{C_{13}}{C_{13} + C_{44}} \right) B_i I_1(1) - 6B_i I_{12} + \frac{D}{4} I_1(3) \]  

(A9)

\[ P_{12} = \left( \frac{C_{13}}{C_{13} + C_{44}} \right) B_i I_1(1) - 2B_i I_{12} - \frac{D}{4} I_1(3) \]  

(A10)

\[ P_{13} = 2B_i \left[ -3I_1(1) + v_i^2 I_2(1) + 4I_{12} \right] \]  

(A11)
\[ P_{33} = 4\nu_1^2 B \left[ 3I_1(1) - \left( \frac{C_{11}}{C_{13} + C_{44}} \right) I_2(1) - 4I_{12} \right] \quad (A12) \]

\[ P_{44} = -B \left[ 3 + \nu_1^2 \left( 3 + \frac{C_{44}}{C_{13} + C_{44}} \right) \right] I_1(1) + \frac{B\nu_1^2}{2} \left( \frac{C_{13}}{C_{13} + C_{44}} \right) I_2(1) + 4B(1 + \nu_1^2)I_{12} + \frac{D}{8}\nu_3^2 I_z(3) \]

\[ (A13) \]

\[ \nu_1 = \nu_2 = \left( \frac{C_{11}}{C_{33}} \right)^{1/4}, \quad k_1 = k_2 = 1 \]

\[ (A14) \]

\[ B_1 = B_2 = -\nu_1 \frac{C_{13} + C_{44}}{16\pi C_{11} C_{44}} \]

\[ (A15) \]

where the integrals \( I_1, I_2 \) and \( I_{12} \) are recalled hereafter (see details in Withers, 1989):

\[ I_1(i) = 2\pi\nu \int_0^1 \frac{(1-x^2)dx}{x^2(1-\nu_1^2) + \nu_1^2} \]

\[ I_2(i) = \frac{4\pi}{\nu_1} \int_0^1 \frac{x^2 dx}{x^2(1-\nu_1^2) + \nu_1^2} \]

\[ I_{12} = \frac{3\pi\nu_1}{2} \int_0^1 \frac{(1-x^2)dx}{x^2(1-\nu_1^2) + \nu_1^2} \]

\[ (A16) \]

\[ (A17) \]

\[ (A18) \]

If \( \nu_1^2 < 1 \)

\[ I_1(i) = \frac{2\pi G}{\nu_1} \left[ \arccos(\nu_1)(1 + G^2) - G \right], \quad G = \frac{\nu_1}{\sqrt{1-\nu_1^2}} \]

\[ (A19) \]

\[ I_2(i) = \frac{4\pi G^2}{\nu_1^3} \left[ 1 - G \arccos(\nu_1) \right] \]

\[ (A20) \]

If \( \nu_1^2 > 1 \)

\[ I_1(i) = \frac{2\pi F}{\nu_1} \left[ \text{arcosh}(\nu_1)(1 + F^2) + F \right], \quad F = \frac{\nu_1}{\sqrt{\nu_1^2 - 1}} \]

\[ (A21) \]

\[ I_2(i) = \frac{4\pi G^2}{\nu_1^3} \left[ F \arccos(\nu_1) - 1 \right] \]

\[ (A22) \]

If \( \nu_1^2 = 1 \)
813 \quad I_1(i) = I_2(i) = \frac{4\pi}{3} \quad (A23)

814 \quad \text{If } 0 < v_1 < 1

815 \quad I_{12} = \frac{3\pi G}{2v_1} \left[ -\frac{5G}{3} - G^2 + (G^4 + 2G^2 + 1)\arccot(G) \right] \quad (A24)

816 \quad \text{If } v_1 > 1

817 \quad I_{12} = \frac{3\pi F}{2v_1} \left[ \frac{5F}{3} - F^3 + (1 - F^2)^2 \arccot(F) \right] \quad (A25)

818 \quad \text{If } v_1 = 1

819 \quad I_{12} = \frac{4\pi}{5} \quad (A23)

820

821 \textbf{Appendix B. Definition and properties of tensor basis } \mathbf{T}

822 T-basis is often introduced to simplify the tensor calculus involving fourth-rank tensors

823 associated with transversely isotropic elastic symmetry (e.g., Walpole, 1981). The objective of

824 this section is twofold: (a) to provide the definition of T-basis and (b) to describe the tensorial

825 operations used in this work, that are all performed in terms of combinations of tensorial

826 components of this particular T-basis.

827 \textit{a- Definition}

828 The tensor basis } \mathbf{T} \text{ is constituted in six tensors or components } (T^1, T^2, T^3, T^4, T^5, T^6) 

829 given by :

830 \quad T^1_{ijkl} = \theta_j \theta_k \delta_{ij}, \quad T^2_{ijkl} = (\theta_i \theta_j + \theta_j \theta_k - \theta_k \theta_i) / 2, \quad T^3_{ijkl} = \theta_{ij} m_k m_l 

831 \quad T^4_{ijkl} = m_i m_j \theta_{kl}, \quad T^5_{ijkl} = (\theta_{ik} m_j m_l + \theta_{ij} m_k m_l + \theta_{jk} m_i m_l + \theta_{jl} m_i m_k) / 4 

832 \quad T^6_{ijkl} = m_i m_j m_k m_l 

833 \text{where } \theta_{ij} = \delta_{ij} - m_i m_j \text{ and } m = m_1 x_1 + m_2 x_2 + m_3 x_3 \text{ is an unit vector along the axis of}

834 transverse symmetry. In our case, this axis is chosen such as: } \mathbf{m} = m_3 \mathbf{x}_3 \text{. These tensors form a

835 closed algebra with respect to the operation of multiplication (contraction over two indices).}
From (B1) and considering the choice \( m = m_3x_3 \), the non-zero components of tensors \((T^1, T^2, T^3, T^4, T^5, T^6)\) are:

\[
T^1_{1111} = T^1_{2222} = T^1_{1122} = T^1_{2211} = 1, \\
T^2_{1212} = T^2_{2121} = T^2_{2112} = T^2_{1111} = T^2_{1122} = -T^2_{2212} = -T^2_{2221} = \frac{1}{2}, \\
T^3_{1133} = T^3_{2233} = 1, \\
T^4_{3311} = T^4_{3322} = 1, \\
T^5_{1313} = T^5_{2323} = T^5_{3123} = T^5_{3223} = T^5_{3131} = T^5_{3232} = \frac{1}{4}, \\
T^6_{3333} = 1. \\
(B2)
\]

\[b-\text{ Properties}\]

If two tensors \( X \) and \( Y \) are given in the \( T \)-basis

\[
X = \sum_{i=1}^{6} x_i T^i, \quad Y = \sum_{i=4}^{6} y_i T^r \\
(B3)
\]

then the contraction of these tensors over two indices, \( X:Y \), in the same basis (tensor with \( ijk \) components equal to \( X_{ijmn}Y_{mnkl} \)) is:

\[
X : Y = (2x_1y_1 + x_3y_4)T^1 + x_2y_2T^2 + (2x_4y_1 + x_3y_6)T^3 + \\
+ (2x_4y_1 + x_3y_4)T^4 + \frac{1}{2} x_4y_5T^5 + (x_6y_6 + 2x_4y_3)T^6 \\
(B4)
\]

Inverse tensor \( X^{-1} \) defined by \( X:X^{-1}X = J \) (or \( X^{-1}_{ijmn} X_{mnkl} = X_{ijmn}X_{mnkl} = J_{ijkl} \)) is given by:

\[
X^{-1} = \frac{x_6}{2\Delta}T^1 + \frac{x_1}{x_2}T^2 - \frac{x_3}{\Delta}T^3 - \frac{x_4}{\Delta}T^4 + \frac{4}{x_3}T^5 + \frac{2x_1}{\Delta}T^6 \\
(B5)
\]

with \( \Delta = 2(x_1x_6 - x_3x_4) \). The fourth-rank unit (symmetrical) tensor \( J \) can be written in the same basis as follows:

\[
J_{ijkl} = \frac{1}{2}(\delta_{ik}\delta_{jl} + \delta_{ij}\delta_{kl}) = \frac{1}{2} T^1_{ijkl} + T^2_{ijkl} + 2T^5_{ijkl} + T^6_{ijkl} \\
(B6)
\]
The components of a transversely isotropic fourth-rank tensor in the T-basis \( T^r \) are linked to their classical components \( X_{ijkl} \) as follows:

\[
x_1 = \frac{1}{2} (X_{1111} + X_{1122}), \quad x_2 = 2X_{1212}, \quad x_3 = X_{1133},
\]

\[
x_4 = X_{3311}, \quad x_5 = 4X_{1313}, \quad x_6 = X_{3333}
\]

(B7)

The inverted relationships are:

\[
X_{1111} = \frac{1}{2} (2x_1 + x_2), \quad X_{1212} = \frac{1}{2} x_2, \quad X_{1133} = x_3,
\]

\[
X_{3311} = x_4, \quad X_{1313} = \frac{1}{4} x_5, \quad X_{3333} = x_6
\]

(B8)

Acknowledgments. This work was initiated from discussions with Maria Zamora who the authors appreciate greatly. We thank the French National Agency for Radioactive Waste Management (Andra), TAPSS 2000 research program (Present and past transfers in a sedimentary aquifer – aquitard system: a 2000 meter deep drill-hole in the Mesozoic of the Paris Basin) and NEEDS-MIPOR for their financial supports.

References


Table captions

Table 1. Mineral composition and petrophysical properties of the clay-rock sample EST05-709 used in this study (from Comparon, 2005).

Hg-porosity is the porosity measured by mercury intrusion. Grain density was determined by helium pycnometry.

Table 2. Geometrical and mineralogical features of the mineral map used in this work (clay-rock sample EST05-709).

Table 3. Input data used to calculate the effective elastic properties of sample EST09-709.

Table 4. Calculated values of effective undrained elastic constants for two dominant clay fractions ($\bar{\phi}_c=0.504$ and $\bar{\phi}_c=0.559$) are compared with dynamic ultrasonic measurements acquired from sample EST05-709.

Table 5. Predicted elastic constants of the clay matrix, $C_{11}$ and $C_{12}$, which provide the best fit between the experimental data and theoretical calculations. The dominant clay fraction $\bar{\phi}_c$ is 0.569 and the values of $C_{13}, C_{33}$ and $C_{44}$ are held constant and correspond to the lower bounds (soft clay matrix) in Table 3. The values of the modelled effective properties and experimental data are also given.

Figure captions

Figure 1. Microstructure of clay-rocks at various scales (modified from Ulm et al. 2005; Sarout and Guéguen, 2008).

Figure 2. Flow-chart of the local clay theory (LCT) used in this work.

Figure 3. Extraction of the mineral map (sample EST05-709) used in this work.

Figure 4. Local clay fraction distribution, $\mu(\phi_c,L)$, calculated for different sizes of measurement cell $L$.

Figure 5. Local percolation probability, $\lambda(\phi_c,L)$, calculated for different sizes of measurement cell $L$. 
Figure 6. Total fraction of percolating cells of size $L$, $p(L)$. For sake of clarity, a polynomial fit is also provided. To use criterion (15), the tangent crossing the curve at the inflexion point is shown.

Figure 7. Local distributions, $\mu(\phi_c, L^*)$ and $\lambda(\phi_c, L^*)$ for $L^*=55$ pixels. The fits used for the calculations of effective properties are also shown (see Eqs. (42) and (43)).

Figure 8. Gaussian functions used in the sensitivity analysis.

Figure 9. Normalized deviation parameters $\Delta_{ij}$ as a function of standard deviation $\sigma_1$ of the Gaussian distribution $\mu(\phi_c, L^*)$.

The $\Delta_{ij}$ values associated with the soft and hard clay matrices are denoted by dashed and solid lines, respectively.

The grey and black lines correspond to $\sigma_1^+=0.5$ (50%) and $\sigma_1^-=0.6$ (60%), respectively.

Figure 10. a: Normalized deviation parameters $\Delta_{ij}$ as a function of parameter $\sigma_1^+$. b: asymmetric local clay fraction distributions $\mu$. In all cases, parameter $\sigma_1^+$ was held constant and equal to 0.05. For comparison, in all cases, the $\Delta_{ij}$ parameters were calculated for the same mean clay fraction, $\phi_c^+$, equal to 60%.

Figure 11. Influence of local clay connectivity parameter values of local distributions $\mu$ and $\lambda$ used in the sensitivity analysis. a: variation range of parameter $\phi_2$ (parameter $\sigma_2$ is held constant at 0.05); b: variation range of parameter $\sigma_2$ (parameter $\phi_2$ is held constant at 0.6, i.e., the value of peak-mean clay fraction $\bar{\phi}_c^2$). The particular value of $\phi_2=0.6(60%)=\bar{\phi}_c^2$ is denoted by a vertical dashed line.

Figure 12. Normalized deviation parameters $\Delta_{ij}$ as a function of parameter $\phi_2$ of the step distribution $\lambda(\phi_c, L^*)$. The parameter $\sigma_2$ is held constant at 0.05. The $\Delta_{ij}$ values associated with the soft and hard clay matrices are denoted by dashed and solid lines, respectively. The grey and black lines correspond to $\sigma_1=0.5$ and $\sigma_1=0.15$, respectively ($\sigma_1$: standard deviation of the Gaussian distribution $\mu$). The particular value of $\phi_2=0.6(60%)=\bar{\phi}_c^2$ is denoted by a vertical dashed line.

Figure 13. Normalized deviation parameters $\Delta_{ij}$ as a function of parameter $\sigma_2$ of the step distribution $\lambda(\phi_c, L^*)$. The parameter $\phi_2$ is held constant $\phi_2=0.6(60%)=\bar{\phi}_c^2$. The $\Delta_{ij}$ values associated with the soft and hard clay matrices are denoted by dashed and solid
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Hg-porosity is the porosity measured by mercury intrusion. Grain density was determined by helium pycnometry.

<table>
<thead>
<tr>
<th>Depth (m)</th>
<th>Quartz (%)</th>
<th>Carbonates (%)</th>
<th>Clay minerals (%)</th>
<th>Smectite (%)</th>
<th>Kaolinite (%)</th>
<th>Chlorite (%)</th>
<th>Illite-smectite (%)</th>
<th>Hg-Porosity (%)</th>
<th>Bulk density (g.cm$^{-3}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>494.76</td>
<td>15</td>
<td>20</td>
<td>65</td>
<td>0</td>
<td>20</td>
<td>30</td>
<td>15</td>
<td>12.4</td>
<td>2.32</td>
</tr>
</tbody>
</table>

Table 2. Geometrical and mineralogical features of the mineral map used in this work (clay-rock sample EST05-709).

<table>
<thead>
<tr>
<th>Resolution ($\mu$m)</th>
<th>Total number of pixels</th>
<th>Dimensions (pixels)</th>
<th>Dimensions ($\mu$m)</th>
<th>Clay minerals (%)</th>
<th>Quartz (%)</th>
<th>Calcite (%)</th>
<th>Others (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>2</td>
<td>384 000</td>
<td>250 x 1536</td>
<td>500 x 3072</td>
<td>50.44</td>
<td>13.83</td>
<td>24.99</td>
<td>10.74</td>
</tr>
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</table>
Table 3. Input data used for the calculations of effective elastic properties of sample EST09-709.

<table>
<thead>
<tr>
<th></th>
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<th></th>
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</thead>
<tbody>
<tr>
<td>Bulk modulus $K_s$</td>
<td>56.3</td>
<td>$C_{11}$</td>
<td>14.3</td>
<td>43.0</td>
<td></td>
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<tr>
<td></td>
<td></td>
<td>$C_{12}$</td>
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<td>17.0</td>
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<tr>
<td>Shear modulus $G_s$</td>
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<td>$C_{13}$</td>
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</tr>
<tr>
<td></td>
<td></td>
<td>$C_{33}$</td>
<td>8.9</td>
<td>11.6</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>$C_{44}$</td>
<td>2.8</td>
<td>2.8</td>
<td></td>
<td></td>
</tr>
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</table>

Table 4. Calculated values of effective undrained elastic constants for two dominant clay fractions ($\bar{\phi}_c$ =0.504 and $\bar{\phi}_c$ =0.569) are compared with dynamic ultrasonic measurements acquired on sample EST05-709.

<table>
<thead>
<tr>
<th>Elastic constants</th>
<th>Experiment [GPa]</th>
<th>Model $\bar{\phi}_c$ =0.504 [GPa]</th>
<th>Model $\bar{\phi}_c$ =0.569 [GPa]</th>
</tr>
</thead>
<tbody>
<tr>
<td>$C_{11}$</td>
<td>27.3</td>
<td>37.4</td>
<td>30.6</td>
</tr>
<tr>
<td>$C_{12}$</td>
<td>13.5</td>
<td>10.9</td>
<td>9.1</td>
</tr>
<tr>
<td>$C_{13}$</td>
<td>*</td>
<td>8.5</td>
<td>6.5</td>
</tr>
<tr>
<td>$C_{33}$</td>
<td>*</td>
<td>29.9</td>
<td>22.6</td>
</tr>
<tr>
<td>$C_{44}$</td>
<td>*</td>
<td>10.6</td>
<td>7.9</td>
</tr>
</tbody>
</table>

*: not measured
Table 5. Predicted elastic constants of clay matrix, $C_{11}$ and $C_{12}$, which provide the best fit between experimental data and theoretical calculations. The dominant clay fraction $\bar{\phi}_c$ is 0.569 and the values of $C_{13}$, $C_{33}$ and $C_{44}$ are held constant and correspond to the lower bounds (soft clay matrix) in Table 3. The values of the modelled effective properties and experimental data are also given.

<table>
<thead>
<tr>
<th></th>
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<tbody>
<tr>
<td>$C_{11}$</td>
<td>12.9</td>
<td>27.3</td>
<td>27.3</td>
</tr>
<tr>
<td>$C_{12}$</td>
<td>8.6</td>
<td>13.5</td>
<td>13.5</td>
</tr>
<tr>
<td>$C_{13}$</td>
<td>2.7</td>
<td>6.4</td>
<td>*</td>
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<tr>
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<td>8.9</td>
<td>22.4</td>
<td>*</td>
</tr>
<tr>
<td>$C_{44}$</td>
<td>2.8</td>
<td>7.5</td>
<td>*</td>
</tr>
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*: not measured
Figure 1. Microstructure of clay-rocks at various scales (modified from Ulm et al. 2005; Sarout and Guéguen, 2008).
Figure 2. Flow-chart of the local clay theory (LCT) used in this work.

Map partitionning

« Local simplicity » assumption
⇒ calculations of
- local functions $\mu(\phi_c, L)$, $\lambda(\phi_c, L)$
- $p(L)$ and consequently the critical length $L^*$

Calculation of effective properties of

percolating cells of size $L^*$
Isotropic sphere in TI clay matrix

blocking cells of size $L^*$
TI sphere of clay in isotropic matrix

Calculation of the effective elastic properties of the clay-rock map seen as a multi-phase composite
Figure 3. Extraction of the mineral map (sample EST05-709) used in this work.

Figure 4. Local clay fraction distribution, $\mu(\varphi_c, L)$, calculated for different sizes of measurement cell $L$. 

\[ \mu(\varphi_c, L) \]

$\varphi_c = 50.44\%$
Figure 5. Local percolation probability, $\lambda(\varphi_c, L)$, calculated for different sizes of measurement cell $L$.

Figure 6. The total fraction of percolating cells of size $L$, $p(L)$. For sake of clarity, a polynomial fit is also indicated. To use criterion (15), the tangent crossing the curve at the inflexion point is shown.
Figure 7. Local distributions, \( \mu(\varphi_c, L^*) \) and \( \lambda(\varphi_c, L^*) \) for \( L^*=55 \) pixels. The fits used for the calculations of effective properties are also shown (see Eqs. (42) and (43)).

Figure 8. Gaussian functions used in the sensitive analysis.
Figure 9. Normalized deviation parameters $\Delta_{ij}$ as a function of standard deviation $\sigma_i$ of the Gaussian distribution $\mu(\varphi_i, L^*)$.

The $\Delta_{ij}$ values associated with the soft and hard clay matrices are denoted by dashed and solid lines, respectively.

The grey and black lines correspond to $\varphi_1 = 0.5$ (50%) and $\varphi_2 = 0.6$ (60%), respectively.
Figure 10. a: Normalized deviation parameters $\Delta_{ij}$ as a function of parameter $\sigma_i^-$. 

b: asymmetric local clay fraction distributions $\mu$. In all cases, parameter $\sigma_i^+$ was held constant and equal to 0.05. For comparison, in all cases, the $\Delta_{ij}$ parameters were calculated for the same mean clay fraction, $\bar{\varphi}_c$, equal to 60%.
Figure 11. Influence of local clay connectivity parameter values of local distributions $\mu$ and $\lambda$, used in the sensitivity analysis. a: variation range of parameter $\phi_2$ (parameter $\sigma_2$ is held constant at 0.05); b: variation range of parameter $\sigma_2$ (parameter $\phi_2$ is held constant at 0.6, i.e., the value of peak-mean clay fraction $\bar{\phi}_c^2$). The particular value of $\phi_2=0.6(60%)=\bar{\phi}_c^2$ is denoted by a vertical dashed line.
Figure 12. Normalized deviation parameters $\Delta_{ij}$ as a function of parameter $\phi_2$ of the step distribution $\lambda(\phi_2; L_\mu)$. The parameter $\sigma_2$ is held constant at 0.05. The $\Delta_{ij}$ values associated with the soft and hard clay matrices are denoted by dashed and solid lines, respectively. The grey and black lines correspond to $\sigma_1=0.05$ and $\sigma_1=0.13$, respectively ($\sigma_1$: standard deviation of the Gaussian distribution $\mu$). The particular value of $\phi_2=0.6(60\%)=\phi_c^{2\%}$ is denoted by a vertical dashed line.
Figure 13. Normalized deviation parameters \( \Delta_{ij} \) as a function of parameter \( \sigma_2 \) of the step distribution \( \lambda(\varphi_c; L^*) \). The parameter \( \varphi_2 \) is held constant \( \varphi_2 = 0.6(60\%) = \varphi_{c}^2 \).

The \( \Delta_{ij} \) values associated with the soft and hard clay matrices are denoted by dashed and solid lines, respectively.

The grey and black lines correspond to \( \sigma_1 = 0.05 \) and \( \sigma_1 = 0.13 \), respectively (\( \sigma_1 \): standard deviation of the Gaussian distribution \( \mu \)).