Negative Unjacketed Pore Modulus in Limestones? Critical Examination of a Peculiar Laboratory Observation

Wenhui Tan¹,², Tobias M. Müller¹,³, and Jing Ba¹

¹School of Earth Sciences and Engineering, Hohai University, Nanjing, China, ²Department of Exploration Geophysics, Curtin University, Perth, WA, Australia, ³Department of Seismology, Centro de Investigación Científica y de Educación Superior de Ensenada, Ensenada, Mexico

Abstract There is recent experimental evidence that the effective pressure coefficient for pore volume exceeds unity in limestones with dual porosity structure. Within the realm of linear poroelasticity, this means that the unjacketed pore modulus is negative. This would be a rather unusual rock behavior since it implies that the pore volume increases for a positive pressure increment during hydrostatic compression. Generally, any deviation of the unjacketed pore modulus from strictly positive mineral bulk modulus means that the deformation is inhomogeneous. We critically examine these observations by attempting to reconcile them with the poroelastic constitutive equations of micro-inhomogeneous rocks that encompass inhomogeneous deformations. Consistency checks among measured poroelastic constants reveal that the measurement accuracy of pore volume changes concurring with the deformation experiments is not as high as desired. However, assigning moderate variations to the measured pore volume changes either due to random or systematic measurement errors, we show that the experimental observations are indeed reconcilable with the constitutive poroelastic equations. Hence, the negative unjacketed pore modulus observed is deemed plausible though overestimated. Within the poroelasticity framework for micro-inhomogeneous rocks, a negative unjacketed pore modulus is the result of a partial localization of elastic strain energy in the fluid space. This in turn could be caused by dissolved calcite particles that block thin pore throats and thus generate temporary fluid pressure compartmentalization. A far-reaching consequence of a negative unjacketed pore modulus is that description of the undrained bulk modulus in rocks with dual porosity structure is more complicated than previously thought.

1. Introduction

Since the groundbreaking works of Biot (1941, 1962) the description of porous, fluid-saturated rocks as poroelastic composites has been of great interest in geomechanics and related disciplines in the Earth sciences. One particular and controversial aspect of the macroscopic theory of poroelasticity is its applicability to so-called micro-inhomogeneous rocks. The latter are rocks that can be conceptualized as macroscopically homogeneous, even though they are heterogeneous at pore-scale, for example, due to the presence of different minerals constituting the rock frame. Biot and Willis (1957) provided generic recipes how to measure the poroelastic constants appearing in the Biot theory. Early attempts to measure poroelastic constants from static deformation experiments in the laboratory have been reported by Geertsma (1957). In these works, in agreement with the theory of Gassmann (1951), the number of independent poroelastic constants could be reduced by one based on the assumption that the deformation proceeds at macro- and micro-scale in a homogeneous manner. Later, Brown and Korringa (1975) showed that in micro-inhomogeneous rocks one additional poroelastic constant is needed. In fact, Brown and Korringa argued that the unjacketed pore modulus is different from the unjacketed bulk modulus in micro-inhomogeneous rocks. This difference has been examined in a modeling study by Berge and Berryman (1995). Interestingly, they found that certain micro-geometries would even give rise to a negative unjacketed pore modulus. For a single elastic continuum, bulk compressibilities are strictly positive and only so-called linear compressibility’s can become negative in relatively rare materials (Cairns & Goodwin, 2015). Since, poroelasticity can be conceived as a theory of two interwoven continua (Müller & Sahay, 2019), such a restriction does not apply. This means that, in principle, a negative bulk modulus is possible.
To understand the issue around the unjacketed pore modulus, let us introduce the four moduli that can be defined if the confining pressure ($p_c$) and fluid pressure ($p_d$) can be independently varied in a deformation experiment, in which the bulk ($V_b$) and the pore ($V_p$) volume changes are measured. Following the notation of Wang (2000), the drained-frame modulus $K_0$, the unjacketed bulk modulus $K_s'$, drained pore modulus $K_p$, and the unjacketed pore modulus $K_\eta$ are

$$\frac{1}{K_0} = -\frac{1}{V_{b0}} \frac{\partial V_b}{\partial p} \bigg|_{p'=0},$$

$$\frac{1}{K_s} = -\frac{1}{V_{b0}} \frac{\partial V_b}{\partial p'} \bigg|_{p''=0},$$

$$\frac{1}{K_p} = -\frac{1}{V_{p0}} \frac{\partial V_p}{\partial p'} \bigg|_{p''=0},$$

$$\frac{1}{K_\eta} = -\frac{1}{V_{p0}} \frac{\partial V_p}{\partial p'} \bigg|_{p''=0},$$

where the subscript 0 refers to the initial (unperturbed) quantity. $K_0, K_s'$, and $K_p$ are often measured (Coyner, 1984; Fatt, 1959; Hughes & Cook, 1953; Newman, 1973; Nur & Byerlee, 1971; Sampath, 1982; Vander Knapp, 1959; Zimmerman et al., 1986). However, there are only few measurements of $K_\eta$ (e.g. Tarokh et al., 2018). This is because pore volume changes are difficult to measure in a hydrostatic experiment. This sparsity of experimental data makes it difficult to verify the assumption of homogeneous deformation and thus check the relations among the poroelastic constants.

According to Brown and Korringa (1975) for micro-homogeneous rocks one should expect

$$K_\eta = K_s' = K_s,$$

where $K_s$ is the bulk modulus of the solid component. Conversely, if the rock is micro-inhomogeneous, $K_s'$ will not be equal to $K_\eta$ and also be different from $K_s$. Figure 1 illustrates the relative volume changes required so that the unjacketed pore modulus becomes negative.

Let us assume, that there is a positive increment in confining pressure in this unjacketed experiment. Then, in order to obtain $K_\eta<0$ the deformation is such that the pore volume enclosed in the representative volume element (RVE) exceeds the pore volume prior to deformation.

As of today, it remains unclear what type of rocks and what type of micro-inhomogeneity cause a violation of Equation 5 and renders a negative unjacketed pore modulus. To further our understanding about this issue, we examine recently published experimental observations of Wang et al. (2018). While they did not directly measure the unjacketed pore modulus $K_\eta$, their measurements of the effective pressure coefficient for the pore volume provide an indirect means to delineate $K_\eta$. Therefore, in the next section we will explain the close connection between these two quantities. In Section 3 we provide the necessary theoretical background and explain our notion of micro-inhomogeneous rocks embedded in the poroelasticity framework of Müller and Sahay (2016). Based on this framework, in Section 4 we analyze the experimental data of Wang et al. (2018). We estimate poroelastic end-member properties and check for consistency among the measured quantities. In Section 5 we interpret and discuss the condition and potential reason for the negative unjacketed pore modulus inferred for some of the limestones.
Let us outline the rationale of our data analysis. We focus on recently reported measurements of poroelastic constants in a selection of limestone samples performed by Wang et al. (2018). They report direct measurements of the drained pore modulus $K_p$ and the effective pressure coefficient for the pore volume $\beta$ (Table 1).

Further below we will assess the measurements of Wang et al. (2018) in more detail. In our context, their measurements are of interest since they enable us to infer the unjacketed pore modulus based on the known poroelastic relation (Berryman, 1992)

\[
\eta = -p \frac{1}{K_p}.
\]

We note that this equation has been derived on the basis of linear poroelastic material behavior. Since the validity of Equation 6 is critical to our analysis, we will re-derive it in Section 3 based on poroelasticity.

### Table 1

| Sample   | $\eta_0$ (%) | $\beta$ | $\alpha^*$ | $K_p$ (GPa) | Mineral composition               | $K_s$ (GPa)$^a$ |
|----------|--------------|---------|------------|-------------|-----------------------------------|----------------|
| Purbeck (P) | 13.6        | 1.45    | 0.6        | 1.25        | 80% calcite, 20% quartz            | 64.67          |
| Thala (T)  | 15.7        | 1.5     | 0.83       | 0.77        | 78% calcite, 22% dolomite          | 75.15          |
| Indiana (I)| 18.1        | 2       | 0.8        | 0.59        | 100% calcite                       | 74.8           |
| Leitha (L) | 29.2        | 0.2     | 0.7        | 1.43        | 100% calcite                       | 74.8           |

$^a$The solid phase bulk modulus is computed using the Voigt-Reuss-Hill (VRH) average.
framework for micro-inhomogeneous rocks of Müller and Sahay (2016). We further note that the assumption of linear material behavior will be adopted throughout this paper.

The peculiarity of the Wang et al. (2018) measurements is that they find that $\beta$ exceeds unity. In fact, they claim that this is the first clear experimental observation for $\beta > 1$. This, in turn, implies that the unjacketed pore modulus has to be negative (since $K_\eta$ is strictly positive). Thus, one is left with the inequality

$$K_\eta < 0 < K_s.$$  

This means, that not only the unjacketed pore modulus is different from the solid phase bulk modulus, in addition $K_\eta$ is negative. These findings might seem counter-intuitive since some of the limestone samples consist of a pure calcite solid phase. Thus, the nature of micro-inhomogeneity cannot be related to a variable mineralogy. Wang et al. (2018) relate this unusual behavior to the presence of micro-porosity. We critically examine the plausibility of the negative pore modulus implied by the measurements of Wang et al. (2018). To do so, we make use of known interrelations between poroelastic constants involving the effective pressure coefficients for the bulk and pore volume and porosity. By constructing plausible modeling scenarios, we try to reconcile the measurement results of Wang et al. (2018) with the poroelastic constitutive equations for micro-inhomogeneous rocks (Müller & Sahay, 2016).

3. Theoretical Background

3.1. The Notion of Micro-Inhomogeneous Rocks

Even though the notion of micro-inhomogeneous rocks is sometimes used in analyses of rock deformation, more often than not a precise definition of this concept is not given. We define micro-inhomogeneity as follows.

First, let us assume that there exists a macroscopic description of the rock, in which all the pore-scale complexities appear in some averaged form. The theory of poroelasticity is such a macroscopic description. This means, that there has to be a scale separation between the pore-scale and the macro-scale. From a laboratory point of view, the macro-scale is the size of a rock sample, typically on the order of a few centimeters. Clearly, if a macroscopically homogeneous rock sample is analyzed through microscopic imaging techniques, one can observe the geometrical complexities at the level of grains and pore spaces: Irregularly shaped grains with sharp corners and pore spaces of complex and erratic shapes rendering pore boundaries complex and irregular surfaces. These are geometrical micro-inhomogeneities.

Elastic deformation can proceed in a homogeneous or inhomogeneous manner. By definition, for a homogeneous deformation the strain tensor is constant throughout the whole deformed domain. Correspondingly, an inhomogeneous deformation results in a spatially variable strain field. Typically, one aims to deform a rock sample such that the macroscopic deformation is homogeneous. Only, then the ratio of applied stress and deformation results in a meaningful deformation modulus. However, at pore-scale, the deformation is most likely inhomogeneous, as the pore-scale strain tensor is not a constant unless very idealized porous structures are considered. Thus, one should expect that the variable pore-scale strain enters the macroscopic description of micro-inhomogeneous rocks in some averaged form.

3.2. Poroelastic Constitutive Equations for Micro-inhomogeneous Rocks

The constitutive equations of poroelasticity that can handle micro-inhomogeneity were developed by Müller and Sahay (2016) based on the earlier work of Sahay (2013) and in accordance with the volume averaging framework of poroelasticity (see Müller & Sahay, 2019 for an introduction). If $p^c$ and $p^f$ denote the confining and fluid pressure, respectively, then for isotropic rocks and volumetric deformations, these constitutive equations are

$$-p^c = K_\alpha \delta_{i,j} - \alpha^* p^f.$$  

This equation relates the confining pressure $p^c$ to the volumetric strain $\delta_{i,j}$, the intrinsic permeability $K_\alpha$, and the interfacial tension $\alpha^*$. The equation describes how the fluid pressure changes in response to volumetric strains.
Here, the measures of deformation are the volumetric strain of the solid phase, $s_{jj}$, and the increment of fluid content $\zeta$. The third measure of deformation is the change of porosity, $\eta - \eta_0$. It is constrained by the porosity perturbation equation

$$\eta - \eta_0 = \frac{(1 - \eta_0)\alpha}{K_0}(\rho^f - (\eta_0 + (1 - \eta_0)n)p^f).$$

These constitutive equations contain the following coefficients. $K_0$ is the drained frame bulk modulus and $\alpha^*$ is the effective pressure coefficient. $M^*$ is the fluid storage coefficient and $\alpha$ is the Biot coefficient. $\eta_0$ is the unperturbed porosity and $n$ is as effective pressure coefficient for the porosity.

It is expedient to distinguish between poroelastic end-member constants and lumped constants. End-member constants have clearly identifiable bounds. The five end-member constants pertaining to the isotropic case are the bulk modulus of the solid ($K_s \geq 0$ and fluid $K_f \geq 0$), which are strictly positive, the unperturbed porosity ($0 \leq \eta_0 \leq 1$), the porosity effective pressure coefficient $n$, and decrement of elasticity parameter $\delta_{K_s}$. The bounds for the $n$–parameter have been found in Sahay (2013). In particular, the upper bound is deduced by considering the undrained bulk modulus resulting from an isostrain deformation experiment in which the fluid and solid masses within the RVE are conserved. Then, the maximum possible effective bulk modulus of such a composite medium is the Voigt average and it turns out that the Voigt average is attained if $n = K_s/K_f$. Thus, $0 \leq n \leq K_s/K_f$. The $\delta_{K_s}$ parameter links the drained frame bulk modulus $K_0$ to the solid-phase bulk modulus $K_s$, $K_0 = (1 - \eta_0)(1 - \delta_{K_s})K_s$. Thus, it can be viewed as a measure of decrement of solid phase (mineral) bulk modulus. It is clearly bounded by $0 \leq \delta_{K_s} \leq 1$. The lumped constants in terms of these end-member properties are summarized in Table 2.

### 3.3. Drained Pore Modulus and Unjacketed Pore Modulus

As the drained pore modulus and the unjacketed pore modulus are our main interest in this paper, we elaborate how these moduli are connected with the poroelastic constants following the recipe given in Müller and Sahay (2016). According to, its definition (Equation 3), the drained frame pore modulus $K_p$ involves changes of pore volume. Since the pore volume changes either through a change in the bulk volume ($dV_b$) or through a change in porosity ($d\eta = \eta - \eta_0$), the pore volume change can be expressed as

$$dV_p = \eta_0 dV_b + V_b d\eta.$$  

Thus, in a drained deformation experiment wherein the fluid pressure perturbation is zero, this pore volume change becomes
\[
\frac{\partial V_p}{\partial p^f} \bigg|_{p^f=0} = \eta_0 \frac{\partial V_b}{\partial p^f} \bigg|_{p^f=0} + V_{0b} \frac{\partial \eta}{\partial p^f} \bigg|_{p^f=0}.
\] (12)

The first term on the right-hand side corresponds to the drained frame bulk modulus (see Equation 1). The second term is evaluated using the porosity perturbation Equation 10. Thus, Equation 12 becomes
\[
\frac{1}{K_p} = \frac{1}{V_{0b}} \frac{\partial V_p}{\partial p^f} \bigg|_{p^f=0} = \frac{1}{K_0} \left( 1 + \frac{1}{\eta_0} \frac{\delta_{K_s}}{\eta_0} \right).
\] (13)

yielding the desired expression of the drained pore modulus in terms of the poroelastic end-member properties. This expression also shows that \(K_p\) always is positive.

Subjecting the differential for the pore volume (11) to the hydrostatic condition, \(p^c = p^f\), yields to
\[
\frac{\partial V_p}{\partial p^f} \bigg|_{p^f=0} = \eta_0 \frac{\partial V_b}{\partial p^f} \bigg|_{p^f=0} + V_{0b} \frac{\partial \eta}{\partial p^f} \bigg|_{p^f=0}.
\] (14)

As before, the last term is evaluated using the porosity perturbation Equation 10. Considering the definitions of the bulk moduli we obtain for the unjacketed pore modulus
\[
\frac{1}{K_\eta} = \frac{1}{V_{0b}} \frac{\partial V_p}{\partial p^f} \bigg|_{p^f=0} = \frac{1}{K_0} \left( 1 - \alpha + \frac{1}{\eta_0} (\alpha - \alpha^*) \right).
\] (15)

In terms of the poroelastic end-member properties this becomes
\[
\frac{1}{K_\eta} = \frac{1}{K_s} \left( 1 + \frac{1}{\eta_0} (1-n) \frac{\delta_{K_s}}{1-\delta_{K_s}} \right),
\] (16)

which coincides with Equation 34 in Müller and Sahay (2016). From this equation the condition for a negative \(K_\eta\) is readily obtained. For \(K_\eta < 0\), the expression enclosed in the bracket must be negative. This yields the condition
\[
n > 1 + \frac{\eta_0 (1 - \delta_{K_s})}{\delta_{K_s}},
\] (17)

That is, the effective pressure coefficient for porosity exceeds unity. Since \(n\) is bounded by \(n \delta_{K_s} \leq 1\), the \(n\)-range in which \(K_\eta\) becomes negative is
\[
1 + \frac{\eta_0 (1 - \delta_{K_s})}{\delta_{K_s}} < n \leq \frac{1}{\delta_{K_s}}
\] (18)

We will make use of inequality (18) further below to define a physically plausible range for a negative \(K_\eta\).

We note that a wider upper bound for \(n\) exists, \(n \leq K_s/K_\eta\), implying an obvious extension of inequality (18).

### 3.4. Effective Pressure Coefficient for Pore Volume

Let us understand how the effective pressure coefficient for the pore volume (\(\beta\)) is embedded in the poroelastic constitutive equations for micro-inhomogeneous rocks (Equations 8–10). Since for small deformation the linearized pore volume change given by Equation 11 must be always true, the relative pore volume change is
\[
\left( \frac{dV_p}{V_{0b}} \right) \left( \frac{dV_b}{V_{0b}} \right) \left( \frac{d\eta}{\eta_0} \right) = \left( \frac{dV_b}{V_{0b}} \right) \left( \frac{d\eta}{\eta_0} \right).
\] (19)
The relative bulk volume change follows from the first constitutive Equation 8

\[
\frac{dV_b}{V_{b0}} = -\frac{p^e + \alpha^* p^f}{K_0}.
\]  

(20)

In turn, the relative porosity change follows from Equation 10

\[
\frac{\hat{\rho} \eta}{\eta_0} = \frac{1}{\eta_0} - \frac{\alpha - \eta_0}{K_0} \left( p^e - (\eta_0 + (1 - \eta_0)n)p^f \right).
\]  

(21)

Substituting these relative bulk and porosity changes in Equation 19 for the relative pore volume change and collecting the confining and fluid pressure terms, we obtain

\[
\frac{dV_p}{V_{p0}} = \left( -\frac{1}{K_0} - \frac{1}{\eta_0} \frac{\alpha - \eta_0}{K_0} \right) p^e + \left( \frac{\alpha^*}{K_0} + \frac{1}{\eta_0} \frac{\alpha - \eta_0}{K_0} (\eta_0 + (1 - \eta_0)n) \right) p^f.
\]  

(22)

In the next step, we express \( \alpha \) and \( \alpha^* \) in terms of the endmember properties \( \delta_{K_s} \) and \( n \) (see Table 2) and obtain

\[
\frac{dV_p}{V_{p0}} = -\frac{1}{K_p} \left( 1 + \frac{1}{\eta_0} (1 - \eta_0) \delta_{K_s} \right) p^e + \frac{1}{K_p} \left( \eta_0 + (1 - \eta_0) \delta_{K_s} + \frac{1 - \eta_0}{\eta_0} \delta_{K_s} n \right) p^f.
\]  

(23)

In the first term, on the right-hand side we identify \( K_p \) (Equation 13). In the second term, on the right-hand side we wish to identify \( K_s \) in terms of its endmember properties (Equation 16). To do so, we re-write

\[
X = \frac{\eta_0 - 1 + (1 - \eta_0) \delta_{K_s} + \frac{1 - \eta_0}{\eta_0} \delta_{K_s} (n - 1)}{\eta_0} + \frac{1 + \frac{1 - \eta_0}{\eta_0} \delta_{K_s}}{\eta_0}
\]  

(24)

Pulling out \( -1/K_p \) in Equation 23 and making use of Equation 24 we obtain the final expression for the relative pore volume change

\[
\frac{dV_p}{V_{p0}} = -\frac{1}{K_p} \left( 1 + \frac{1}{\eta_0} (1 - \eta_0) \delta_{K_s} \right) p^e + \frac{1}{K_p} \left( \eta_0 + (1 - \eta_0) \delta_{K_s} + \frac{1 - \eta_0}{\eta_0} \delta_{K_s} n \right) p^f.
\]  

(25)

From Equation 25 the effective pressure coefficient for the pore volume can be read off as

\[
\beta = 1 - \frac{K_p}{K_s}.
\]  

(26)

As expected, this coincides with Equation 6. For the data modeling presented below it will be useful to express the effective pressure coefficient \( \beta \) in terms of the poroelastic endmember properties. Substituting Equations 13 and 16 into Equation 26 we obtain

\[
\beta = 1 - \frac{\eta_0 (1 - \delta_{K_s}) + (1 - n) \delta_{K_s}}{\eta_0 + (1 - \eta_0) \delta_{K_s}}.
\]  

(27)
The condition that $\beta > 1$ means that the expression in the numerator must be negative. Simple algebraic manipulations yield the inequality (17). This means that the condition for a negative unjacketed pore modulus is indeed the same as the condition for the pore volume effective pressure coefficient exceeding unity.

4. Analysis of Experimental Data

4.1. Measured Poroelastic Constants, Uncertainties, and Assumptions

Wang et al. (2018) measure the porosity and the effective pressure coefficient for the pore volume $\beta$ among other quantities. In general, $\beta$ itself varies in the $(p_c, p_f)$ domain. For the reported measurements, $p_c$ ranges from 9 to 18 MPa and $p_f$ ranges from 1.3 to 5.7 MPa, $\beta$ is nearly constant (see Figure 13 in Wang et al., 2018). Therefore, we take the $\beta$-values in Table 1 (which were read off from Figure 13 in Wang et al., 2018) as representative for these pressure conditions. The experimental errors involved in these measurements are somewhat difficult to assess. Wang et al. (2018) mention in this regard that the measured pore volume changes are thought to be accurate for the Indiana (I), Purbeck (P), and Thala (T) samples. Only for the Leitha (L) sample a substantial measurement inaccuracy is expected, but Wang et al. (2018) assert that $\beta$ is less than unity. Wang et al. (2018) verified that the pore volume change is linear with the pressure change (their Figure 14), thereby giving support to a linear poroelastic behavior. The drained pore modulus $K_p$ is inferred from the drained pore compressibility from the slope in the pore volume versus pressure change plot (their Figure 14). They further measure the effective pressure coefficient for the bulk volume ($\alpha^{*}$) based on axial strain measurements. We note that this reasoning implicitly assumes that the rock samples are isotropic. From their Figure 17, we read off the $\alpha^{*}$ values for the same pressure ranges as for $\beta$ (see Table 1). The second last row in Table 1 lists the mineral composition for the four limestones. The I and L samples apparently consist of 100% calcite. The last row in Table 1 shows the solid phase bulk modulus. It has not been measured, however, since the mineralogy of the samples is known, we use the Voigt-Reuss-Hill (VRH) average (Hill, 1952) to evaluate $K_s$.

4.2. Estimating Poroelastic End-Member Properties, Consistency Checks, and Conflicting Results

In a first attempt to model the above observations, we evaluate the poroelastic end-member properties and check for consistency among the measured quantities. These results will be presented in Tables 3–5 and for a quick reference, the main assumptions and findings are given in the footnotes of each Table.

Table 3

| Sample | $K_0$ (GPa) | $\delta K_s$ | $\alpha$ | $\alpha^{*}$ | $n$ |
|--------|-------------|--------------|----------|-------------|-----|
| P      | 8.05        | 0.856        | 0.876    | 1.29        | 1.56 |
| T      | 4.61        | 0.927        | 0.939    | 1.42        | 1.61 |
| I      | 3.12        | 0.949        | 0.958    | 1.92        | 2.24 |
| L      | 4.60        | 0.913        | 0.939    | 0.21        | -0.13 |

Measurements of $\eta_0$, $\beta$, and $\alpha^{*}$ are used. $\alpha^{*}$ values are unphysical. $n\delta K_s$ bound is violated.

Table 4

| Sample | $K_0$ (GPa) | $\delta K_s$ | $\alpha$ | $\alpha^{*}$ | $n$ |
|--------|-------------|--------------|----------|-------------|-----|
| P      | 8.05        | 0.856        | 0.876    | 1.29        | 1.56 |
| T      | 4.61        | 0.927        | 0.939    | 1.42        | 1.61 |
| I      | 3.12        | 0.949        | 0.958    | 1.92        | 2.24 |
| L      | 4.60        | 0.913        | 0.939    | 0.21        | -0.13 |

Measurements of $\eta_0$, $\beta$, and $K_p$ are used. $n\delta K_s$ bound is violated.
First, we only use the experimental data subset {$\eta_0, \beta, \alpha^\star$} to check for consistency and to estimate $K_p$. We further assume that the effective pressure coefficient is the same as the Biot coefficient, $\alpha^\star = \alpha$ (the standard assumption in Biot’s theory valid for micro-homogeneous rocks). This assumption allows us to compute the drained bulk modulus and the decrement of elasticity (second and third rows in Table 3). Substituting these $K_0$ and $\delta K_s$ into Equation 13 yields $K_p$ (fourth row in Table 3). Finally, with $\delta K_s$ and $n$ determined, we solve Equation 27 for the porosity effective pressure coefficient $n$ (second last row in Table 3). We make several observations. Since, it must be always true that $n \geq 0$, the negative value for the L sample is unphysical and points to an inconsistency in the estimates in Table 3. By inspection, we find that the upper bound $\delta \leq K_s n$ does not hold for the other two samples. This also points to a problem with the so-determined constants $\delta K_s$ and $n$ in Table 3. Further, the estimated $K_p$ values significantly exceed the measured ones. From this modeling scenario we infer that using the measured $\alpha^\star$ and/or $\beta$ values results in a mutually inconsistent data set independent of the measured values of $K_p$. This might point to experimental errors associated with $\alpha^\star$ and/or $\beta$.

We do a further, consistency check with the experimental data subset {$\eta_0, K_p, \alpha^\star$} to estimate $\beta$ and $n$. The results are shown in Table 4. Our first observation is that the drained bulk modulus is very low; for example, for the I sample $K_0$ would be comparable to the bulk modulus of water. More importantly, the values of $\alpha^\star$ are all unphysical. For the P, T, and I sample this value exceeds unity and for the L sample it is less than the porosity. Also, the product $n \delta K_s$ is outside the permitted range. This means that, independent of the accuracy of the $\alpha^\star$ measurement, the measured values of $\beta$ and/or $K_p$ does not give us a data set that can be interpreted with linear poroelasticity.

Our last consistency check uses the experimental data subset {$\eta_0, K_p, \alpha^\star$} to estimate $\beta$ and $n$. The results are shown in Table 5. In this scenario all the constants are within the bounds. However, the drained bulk modulus appears to be unreasonably small. It would be also incompatible with the assertion of Wang et al. (2018) that $\beta > 1$ for the T and I samples.

The above calculations demonstrate that there is no internal consistency among the poroelastic constants. More importantly, the poroelastic endmember properties inferred from the measured data violate bounds and are thus deemed unphysical. We conclude, that there are either systematic measurement errors associated with the values listed in Table 1, or that one or more assumptions (linear poroelasticity and isotropy) are violated. In the following, we assume that linear poroelasticity holds and examines how the propagation of measurement error affects the interpretation. Since, our aim is to find out how these measurements can be reconciled with the constitutive equations for micro-inhomogeneous rocks, we construct plausible modeling scenarios by varying some of the measured values within a reasonable range.

### 4.3. Estimating Poroelastic Constants Adopting Meaningful Assumptions for Micro-inhomogeneous Rocks

According to, Wang et al. (2018), one suspected source of the unusual observation $\beta > 1$ for the P, T, and I samples is due to micro-inhomogeneity, which they attributed to the dual porosity character of these
limestones. In the light of the poroelastic constitutive Equations 8–10 the presence of micro-inhomogeneity means that the assumption $\alpha^* = \alpha$ is not valid. Therefore, in our first modeling scenario we will relax this assumption (and discard the measurements of $\alpha^*$). We also noticed from Tables 4 and 5 that the values of the drained bulk modulus become almost unreasonably small when $K_0$ is estimated from the measured drained pore modulus $K_p$. Hence, to start with reasonable estimates of $K_0$, we make use of the drained bulk modulus versus porosity trend observed for a collection of limestones (Figure 2; this data is taken from Table A1).

With these $K_0$ estimates the values of the Biot coefficient $\alpha$ and $K_p$ are determined (Table 6). The consistency checks in Tables 3 and 4 indicate that the direct use of the $\beta$ values result in unphysical predictions. We therefore deduce that the $\beta$-measurements come with some experimental error and instead use model values that deviate from the measured value (the percentage of deviation is given in brackets in Table 6). For P, T and I we find that errors of 40% or less in $\beta$ yield to physically plausible scenarios. Only for L the error in $\beta$ is taken to be larger (300%) in order to obtain a physically plausible case. But this is in line with the anticipated large error in the $\beta$-measurement for the L sample (Wang et al., 2018).

A complementary modeling scenario is to assume that the measured $K_p$-values are in error and hence to use model $K_p$ values. We choose the drained pore modulus such that the drained bulk modulus is consistent with the limestone trend (Table A1). We further honor the experimentally asserted observation that $\beta > 1$ for P, T, and I and $\beta < 1$ for L. Then, the possible range of $n$-values is $1 + \frac{\eta_0}{\delta K_s} (1 - \delta K_s) < n \leq \frac{1}{\delta K_s}$. The resulting range of $\beta$ values is summarized in Table 7. We note that for P and L the respective maximum and minimum $\beta$ values are close to the measured values.

Overall, the following picture emerges. The observations of Wang et al. (2018) can be reconciled with the poroelastic constitutive equations for micro-inhomogeneous rocks only if one accepts that there are measurement errors in the drained pore modulus and the effective pressure coefficient for the pore volume. Given the difficulty to precisely measure the pore volume variation (see Appendix A in Wang et al., 2018), such measurement errors are perhaps expected. We will include these modeling scenarios in our assessment of the unjacketed pore modulus.

5. Discussion: Plausibility of Negative Unjacketed Pore Modulus

5.1. The Inferred Range of $K_p$ Based on the Wang et al. (2018) Data

In Table 8 we summarize the values obtained for the unjacketed pore modulus $K_p$. The values directly inferred from the measurements of Wang et al. (2018) are listed in the second column. For the P, T, and I samples $K_p$ is negative, while for the L sample $K_p$ is positive.

### Table 8

| Sample | $\beta_{MODEL}$ (% error) | $K_p$ (GPa) | $\delta K_p$ | $\alpha$ | $n$ | $\alpha^*$ | $K_p$ (GPa) |
|--------|--------------------------|-------------|-------------|----------|-----|----------|-------------|
| P      | 1.2 (−17)                | 20.94       | 0.625       | 0.676    | 1.33| 0.86     | 4.2         |
| T      | 1.2 (−20)                | 15.43       | 0.756       | 0.79     | 1.3 | 0.99     | 3.04        |
| I      | 1.2 (−40)                | 18.08       | 0.705       | 0.76     | 1.34| 0.95     | 4.32        |
| L      | 0.8 (+300)               | 8.58        | 0.838       | 0.885    | 0.76| 0.74     | 2.83        |

$aK_0$ chosen to follow the limestone trend from Table A1.
To find out, how these measurements can be reconciled with the constitutive equations for micro-inhomogeneous rocks, we constructed plausible modeling scenarios with physically consistent parameter ranges obtained in the previous section. Based on the values from Table 8, the ranges of physically consistent $K_\eta$ values are given in the third column. We observe that these values are less (more negative) than the directly inferred values. A possible reason for this mismatch could be that the pore volume changes have been over-estimated in the experiments. The fourth column gives the $K_\eta$ values for the wider upper bound $n = K_s/K_f$. We observe that these values indeed provide an upper bound for the directly inferred $K_\eta$ values. This means, that the measured values fall into a physically possible range. However, based on the consistency checks among the poroelastic coefficients performed above we think that it is more likely that measurement errors creep into in one or more poroelastic coefficients resulting in over-estimated $K_\eta$ values.

We graphically illustrate our findings in Figure 3, where various $K_\eta$ estimates are shown as a function of porosity. Note that the scales for $K_s$ and $K_\eta$ are different. The directly inferred values are labeled as $\eta^{Exp}$. They increase with porosity and become positive for the L sample. We note that the line connecting the four data points serves only for visual guidance. The physically consistent modeling scenario from Table 8 corresponds to the darker green-shaded area bounded from above by the line $K_s(\delta K_s n = 1)$. This bound does not hold for the L sample, for which our physically consistent modeling scenario (Table 8) yields to a positive range of values, with the upper bound given the bulk modulus of the solid phase $K_s$ (also indicated by a red circle). Opposite to the $\eta^{Exp}$ trend, the line $K_s(\delta K_s n = 1)$ decreases with porosity. This means, that the more negative $K_\eta$ becomes, the smaller are the pore volume changes $dV_p$. If $n$ attained its wider upper bound value $K_s/K_f$ then the largest negative values for $K_\eta$ would be obtained (red squares in Figure 3).

### 5.2. The Fluid Pressure Compartmentalization Hypothesis

What could cause a negative unjacketed pore modulus? Without reference to a specific rock, the regime $n > 1$ generally describes a macroscopic deformation state in which there is a partial localization of the potential energy in the fluid space in some parts of the RVE. Such a localization prevents pore-interface deformation that would occur otherwise to reach a new state of mechanical equilibrium after a change of the confining or fluid pressures. This, in turn, suggests the presence of localized pore-scale fluid pressure compartments within the RVE that counteract the pore-interface deformation. We hypothesize, that such fluid pressure compartments can be generated and sustained over the course of a deformation experiment when local fluid pathways are blocked. We note the blockage does not to be permanent but only over the time span of the experiment.

As it has been observed elsewhere (e.g., Garing et al., 2015), the dissolution process in carbonates is accompanied with the detachment of small carbonate particles. Such particles could accumulate in pore throats and ultimately clog pore throats. This reduces or even blocks the pressure communication and thus leads to localized fluid pressure pockets. Clearly, smaller pore throats provide a favorable condition for such a pressure...
compartmentalization. Another consequence of this carbonate particle accumulation in pore throats is the reduction of the overall permeability, as has been reported by Garing et al. (2015).

As far as the P, T, and I samples are concerned, Wang et al. (2018) find that these carbonate rocks have a dual porosity structure. One porosity system is classified as micro-porosity according to their NMR measurements. Wang et al. (2018) also argue that this micro-porosity is responsible for substantially lower permeability’s in the P, T and I samples, as compared to the permeability of the L sample (which was classified as a single porosity rock). They further argue that this dual porosity structure is the cause of micro-inhomogeneity. Therefore the observations of Wang et al. (2018) are not in contradiction with the hypothesized fluid pressure compartments that lead to a localization of potential energy in the fluid space and hence $n > 1$. If this localization is such that $\frac{\eta}{K_s} \geq n > 1$, the negative values of the unjacketed pore modulus are plausible. Our modeling scenarios indicate that the unjacketed pore modulus is more negative than the measured data suggests. This, in turn, could be the result of an over-estimation of the pore volume changes involved in the measurements.

A far-reaching consequence of a negative unjacketed pore modulus is that the corresponding undrained bulk modulus is beyond the scope of the Gassmann theory. According to Equation (52) of Müller and Sahay the undrained bulk modulus for micro-inhomogeneous rocks is

$$ K_{ud}^{BG} = K_0 + \alpha \left( \eta_0 + \frac{\alpha - \eta_0}{K_s} \right). $$

Since a negative $K_0$ implies an effective pressure coefficient for porosity larger than unity (Equation 17), it follows that $\alpha^* > \alpha$ and hence $K_{ud}^{BG} > K_{ud}^{BG}$. This means, that the undrained bulk modulus would exceed the Gassmann prediction ($K_{ud}^{BG}$) and therefore seismic P-wave velocities would increase. The current data set does not allow us to investigate this aspect in more detail since the undrained bulk modulus has not been measured. However, in agreement with previously reported doubts on the applicability of the Gassmann theory to rocks with complicated micro-structure, in particular carbonates (Adam et al., 2006; Gegenhuber, 2015), our modeling for the three limestones with double-porosity structure also put a question-mark on how to perform fluid-substitution in such rocks.

6. Conclusion

Recent measurements of the effective pressure coefficient for the pore volume in limestones suggest that the unjacketed pore modulus can become negative in limestones. Our aim was to reconcile these observations with the poroelastic constitutive equations of micro-inhomogeneous rocks. Consistency checks among the measured quantities reveal that one has to assume that there are measurement errors involved in the pore volume changes concurring with the deformation experiments. Otherwise, we would have an effective pressure coefficient for the bulk volume larger than unity, which is unphysical regardless of any known, linear poroelasticity theory. It would also imply an almost unreasonably small drained pore modulus. However, if we assign moderate variations to the measured values, either due to random or systematic measurement errors, our modeling shows that the experimental observation can be reconciled with the constitutive poroelastic equations for micro-inhomogeneous rocks. We think, that an overestimation of pore volume change during the deformation experiments is the most likely source of error.

Given that, any deviation of the unjacketed pore modulus from the bulk modulus of the solid phase is a signature of micro-inhomogeneity, one conclusion we draw from these observations is that even an apparently
mono-mineral frame leads to micro-inhomogeneous rock behavior. This is different from the typical notion of micro-inhomogeneous rocks; wherein micro-inhomogeneity is attributed to the presence of more than one mineral phase. Our findings suggest that the concept of micro-inhomogeneity also needs to encompass irregularly shaped grains with sharp corners and pore spaces of complex and irregular surfaces, as one would expect for so-called double porosity rocks. Within this picture, a negative unjacketed pore modulus is the result of a partial and possibly temporary localization of elastic strain energy in fluid space.

Appendix A: Data collection for limestones

| Sample                      | $\eta_0$ (%) | $K_p$ (GPa) | $K_s$ (GPa) | $\alpha$ | Saturation | Reference                  |
|-----------------------------|--------------|--------------|--------------|----------|------------|-----------------------------|
| West China                  | 0.71–2.5     | 52.8–56.1    | 56.3         | 0.0035–0.062 | dry        | Cheng et al. (2019)         |
| Indiana (1)                 | 13           | 22.8         | 71           | 0.68     | dry        | Hart and Wang (1995)        |
| Lavoux                      | 23           | 15           | 77           | 0.81     | drained    | Pimienta et al. (2016)      |
| Savonnieres                 | 29           | 11.6         | 76.8         | 0.85     | drained    | Mikhailsevitch et al. (2019) |
| Chauvigny                   | 16.5         | 16.3         | 52.6         | 0.69     | dry        | Fabre and Gustkiewicz (1997) |
| Lavoux                      | 21.9         | 13.8         | 59.8         | 0.77     | dry        | Fabre and Gustkiewicz (1997) |
| Villeperdue                 | 6.4          | 36           | 61           | 0.41     | dry        | Fabre and Gustkiewicz (1997) |
| Tonnerre                    | 13           | 19.3         | 41.4         | 0.53     | dry        | Fabre and Gustkiewicz (1997) |
| Morawica marble             | 3.4          | 58           | 75           | 0.23     | dry        | Fabre and Gustkiewicz (1997) |
| Louny gaize                 | 26           | 9.2          | 46.4         | 0.80     | dry        | Fabre and Gustkiewicz (1997) |
| Lixhe chalk                 | 42.8         | 3.8          | 42.5         | 0.91     | dry        | Fabre and Gustkiewicz (1997) |
| Lavoux                      | 23           | 9.1          | 53.5         | 0.83     | salt water | Laurent et al. (1993)       |
| Vilhonneur                  | 14           | 20           | 64.5         | 0.69     | salt water | Laurent et al. (1993)       |
| Larrys-Mouchete             | 4.5          | 33           | 50           | 0.34     | salt water | Laurent et al. (1993)       |
| Indiana (2)                 | 18.1         | 14.9         | 74.5         | 0.8      | dry        | Vajdova et al. (2004)       |

Data Availability Statement

All experimental and modeled data can be accessed at https://doi.org/10.5281/zenodo.3934936.

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