Mechanical properties of organic-rich shales based on nanoindentation: A case study of deep shale gas reservoirs in Zigong area, Sichuan Basin

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Abstract. The deep shale gas in the Zigong area of the Sichuan Basin has a wide distribution and has good prospects for exploration, which has become an important replacement area for shale gas exploration and development. In this area, the burial depth of Longmaxi Formation Shale is generally >3500 m. One of the most challenging tasks in the development of this type of reservoir is the accurate prediction of the mechanical properties of organic-rich shale. The application of the mechanical properties of shale is important to the success in multiple aspects of drilling, hydraulic fracturing, and production. Methods such as unconfined/triaxial compression tests with the use of core plugs and logging data interpretation are commonly used to obtain mechanical properties. However, these methods have their own limitations. Thus, to address the difficulty in the measurement of the mechanical properties of shale, the advanced nanoindentation test is introduced and used. Nanoindentation is a technique based on a hard tip, which is pressed into the surface of a sample. The advantage of this test is the use of a sample with a small volume, such as drill cuttings or fragments, which are easily obtained during drilling. The mechanical properties at the nanoscale level, including the hardness, Young’s modulus, and fracture toughness, are measured in the nanoindentation. Moreover, this technique can be used to evaluate the creep behavior of shale. In this study, grid nanoindentation is used to investigate the mechanical properties of drill cutting samples from the Longmaxi Formation Shale in Zigong area, Sichuan Basin. Subsequently, these mechanical parameters are used to evaluate the heterogeneous properties of shale at the nanoscale level. The results can help in analyzing the correlation between rock mechanical properties and shale constituents. This suggests that the nanoindentation test is a useful approach of estimating the elastic and mechanical properties of shale in situations where conventional rock mechanics test data cannot be measured.

1. Introduction
The Longmaxi Formation Shale is one of the largest unconventional shale plays in the Sichuan Basin [1-2]. In the past decade, the primary developed shale reservoir was at the depth of <3500 m [3]. With the progress in the exploration and development of deep shale gas, which has a burial depth of <3500
m, it is found to have a wide distribution and good prospects for exploration in the Sichuan Basin. The typical deep shale area is in Zigong, which has become an important replacement area for shale gas development in the following decades [4]. Horizontal drilling and hydraulic fracturing, which are commonly used in shale development, are used to increase the production from the Longmaxi Formation Shale [3]. Previous experiences show that having a good knowledge of the geomechanical properties of shale is extremely important in the placement and design of horizontal wells and in hydraulic fracturing [5]. The geomechanical properties of shale are primary controlled by mineralogy and structural properties, as well as the in situ stresses and the anisotropic and heterogeneous characters of shale. In a routine laboratory analysis, standard static unconfined/triaxial lab experiments or dynamic field-scale analysis using sonic logs is usually conducted to obtain the necessary rock properties; however, these methods have their own limitations. In the unconfined/triaxial compression tests, obtaining standard-sized core plugs from downhole cores is challenging and expensive. However, the mechanical properties using sonic logs have a high error; thus, to address the difficulty in measuring rock properties, the advanced nanoindentation test is introduced to obtain mechanical parameters [6-8]. Nanoindentation, which only requires a small volume of rock sample, has proven to be successful in petroleum engineering applications for investigating the mechanical properties of shale. Nanoindentation is a technique based on a hard tip, which is pressed into the surface of a sample [9]. Its advantage includes the use of a sample with a small volume, such as drill cuttings or fragments, which are easily obtained during drilling and often cover large intervals of the well trajectory [10-12]. The mechanical properties at the nanoscale level, including the hardness, Young modulus, and fracture toughness, are measured in nanoindentation [10, 13]. This technique can be used to evaluate the creep behavior of shale [8]. However, a single indent or a few indents cannot provide sufficient information about the properties for a heterogeneous shale sample. This is because shale is widely known to be composed of different minerals, including hard and soft minerals [14-17]. To solve this problem, the method of grid indentation was developed to analyze the mechanical properties of composites [16-17, 23-24]. The principle of this method is to perform a large number of indents on a sample surface with specific requirements. This has become an important method for characterizing material properties. Nanoindentation can reflect the mechanical properties of the samples at the nanoscale level [18, 21]. Some researchers have used this data to estimate mechanical properties at the macroscale level. Averaging of the nanoindentation data from a large number of indents is the conventional method used by researchers to obtain macroscale properties [19-20, 25-26]. In this study, the mechanical properties, including the Young modulus and hardness of the shale collected from Longmaxi Formation Shale at Zigong area, were tested by grid nanoindentation. The first part of this study briefly introduces the nanoindentation methods. The second part discusses the results and discussions of nanoindentation tests, including the load-displacement curve analysis, nanoindentation behavior, data analysis and discussion. Finally, conclusions are provided at the end.

2. Nanoindentation methods

2.1. Grid Nanoindentation
Nanoindentation is based on a hard indenter tip with known properties, which is pressed into the sample surface [16-17]. This technique comprises four steps. First, the indenter comes into contact with the sample surface without pre-stress. Then, the indenter is vertically pressed into the sample surface at a constant load rate. Next, the applied load reaches the maximum value and is held for 15 s. Finally, the indenter is lifted away from the sample surface, and the load unloads to 0 [20]. The Berkovich indenter is commonly used in nanoindentation. It is three-sided and has a symmetrical shape. Figure 1(a) shows the indenter for sensing the load and its associated displacement into the sample. Moreover, the load–displacement curve obtained from nanoindentation can be used to calculate the mechanical properties of the sample. Figure 1(b) shows a typical load–displacement curve in nanoindentation. This curve comprises three stages: loading, holding, and unloading. In the
loading stage, which includes the elastic and plastic deformation of sample, the applied load increases with the increase in the penetration depth. In the unloading stage, elastic deformation can be recovered, and plastic deformation can create fracture.

**Figure 1.** Schematic of (a) the indenter loading–unloading process and (b) the nanoindentation load–displacement curve.

The mechanical properties, primarily the hardness and Young modulus, can be obtained from the load–displacement curve. The following parameters are obtained based on the curve: the maximum load $P_{\text{max}}$, the maximum displacement $h_{\text{max}}$, the permanent depth of penetration $h_f$, and the elastic unloading stiffness $S = \frac{dP}{dh}$, which is quantified as the slope of the upper portion of the unloading curve given by the following equation [8, 10]:

$$S = \frac{dP}{dh} = \beta \frac{2 \sqrt{A_c}}{\sqrt{\pi}} E_r$$

where $\beta$ denotes a constant dependent on the geometry of the indenter ($\beta = 1.034$ for a Berkovich indenter); $A_c$ denotes the contact area between the indenter and sample; and $E_r$ denotes the reduced Young modulus, which is given by the following equation [8, 10]:

$$\frac{1}{E_r} = \frac{1-v_i^2}{E_i} + \frac{1-v_r^2}{E_r}$$
where $E$ denotes the Young modulus and $v$ the Poisson ratio of the samples, and $E_i$ and $v_i$ denote the Young modulus and Poisson ratio of the indenter. For a Berkovich indenter, $E_i = 1141$ GPa and $v_i = 0.07$.

The Young modulus $E$ of the sample can then be determined by using the combination of Eq. (1) and Eq. (2). The hardness can be derived from the following equation [8, 10]:

$$H = \frac{P_{\text{max}}}{A_c}$$  \hspace{1cm} (3)

where $P_{\text{max}}$ denotes the maximum load. The contact area $A_c$ is the function of the contact depth $h_c$ [8, 10]:

$$A_c = 24.5h_c^2$$  \hspace{1cm} (4)

There is a relationship between $h_c$ and $h_{\text{max}}$, which is expressed by the following equation [8, 10]:

$$h_c = h_{\text{max}} - \frac{P_{\text{max}}}{\varepsilon S}$$  \hspace{1cm} (5)

where $\varepsilon$ denotes a geometric constant for the indenter ($\varepsilon = 0.75$ for a Berkovich indenter).

Since shales are highly heterogeneous in constituent components which becomes more noticeable at the nanoscale level [14]. To obtain the mechanical properties of shales, grid indentation analysis is used in the test. A large number of indents are performed on the sample surface, with each indent being considered as an independent event [19]. In this study, the Berkovich indenter was used to conduct the experiments.

2.2. Experimental Samples

The samples were drilled from full-sized cores of Longmaxi Formation Shale at Zigong area, Sichuan Basin, at a depth of 3643 m. Nanoindentations were performed on drill cutting samples on two directions: parallel to the bedding and perpendicular to the bedding. Hence, one set of samples were cut perpendicular to the bedding while the other set parallel to the bedding. These two samples are cylinder shale samples with a diameter of 25 mm and a length of 10 mm. Furthermore, another set of drill cutting samples were obtained at the same depth from an adjacent well, which was drilled by using oil-based drilling fluids. Each aliquot was then prepared using standard methodologies as a polished epoxy resin block. In the above three groups of samples, each group contained three samples. Figure 2 shows the samples used in the nanoindentations.
Figure 2. Experimental samples (a) parallel to the bedding and (b) perpendicular to the bedding and (c) the drill cutting sample

The surface smoothness of the sample specimens is extremely important for the accuracy of the results. To minimize the effect of surface roughness on the accuracy of the results, different grit sizes of sandpaper (from 600 to 1200) were used to polish the samples surface. Then, the samples were carefully polished using argon ion machine to meet the surface roughness requirements. The tests were conducted on the center of the sample, with 10 x 10 grid sets and a spacing of 20 μm.

3. Results and discussions

3.1. Mineralogical Composition

The quality fraction of mineral composition and mineral analysis statistics was obtained using energy-dispersive spectroscopy mapping (Table 1). The results show that shale is primarily composed of quartz minerals, plagioclase, carbonate minerals (calcite and dolomite), and clay minerals. Because of the high fractions of hard minerals, especially quartz at a fraction of 51.14 wt%, it can be concluded that the Longmaxi Formation Shale in Zigong area is relatively brittle, as expected, with high elastic modulus and hardness. The total fraction of clay minerals was 15.81 wt%, and the total fractions of dolomite and calcite were 8.88 and 18.37 wt%, respectively. Calcite is considered as a cementing or bonding material that fills the natural pores and cracks of shale. Moreover, a small amount of pyrite (1.85 wt%), a common mineral found in shale, is present.

| Mineral name   | Total clay | Quartz | Plagioclase | Calcite | Dolomite | Pyrite |
|----------------|------------|--------|-------------|---------|----------|--------|
| Fraction (wt%) | 19.63      | 48.43  | 3.87        | 18.17   | 8.02     | 1.88   |
| Average (wt%)  | 15.81      | 51.14  | 3.95        | 18.37   | 8.88     | 1.85   |
| Hardness/GPa   | 1.03       | 4.89   | 3.68        | 2.37    | 1.94     | /      |
| Young modulus/GPa | 12.73  | 65.26  | 53.76       | 59.18   | 43.67    | /      |

3.2. Analysis of Load–Displacement Curve and Nanoindentation Behavior

Figure 3 shows the typical load–displacement curves obtained from the tested samples. These curves present the elastic–plastic deformation during the loading process. Figure 3 (a) shows the typical indentation curves for the sample parallel to the bedding, sample perpendicular to the bedding, and drill cutting sample. From the figure, the sample perpendicular to the bedding has the highest
permanent depth of penetration, whereas the drill cutting samples have the lowest values. This indicates that the oil-based drilling fluids weaken the properties of drill cuttings. In certain indentations, the obtained load–displacement curves are not smooth in certain stages, and some “pop-in” behaviors occur at the loading stage, as shown in Figure 3(b). The main reason for the occurrence of “pop-in” behavior is that the indenter changes the elastic energy of the sample; once the elastic energy increases to a critical value at some point in the contact field [20], plastic deformation occurs with the increasing load. As the load approaches the yield strength of the rock, fractures occur. As the fracture tip begins to expand inward rock, and the process continues until load stops. Moreover, the heterogeneity of the shale could cause the “pop-in” phenomenon.

![Graph showing load-displacement curves](image)

**Figure 3.** Typical nanoindentation load–displacement curves: (a) normal curve, (b) “pop-in” phenomena in loading

Figure 4 shows the scanning electron microscopy images of the grid indentation matrix and single indentation. As can be seen from Figure 4(a), some intrinsic pores and microcracks exist in the
samples, which may result in the failure of indentation. Figure 4(b) shows that the number of microcracks in the central of the indentation is much less than that of the contact area of the indenter edge. The reason for this is that stress concentration occurs in the contact area of the indenter edge and sample surface. Moreover, these fractures have the potential to go along the edges of the existing pores or microcracks.

![Figure 4. Scanning electron microscopy images of the grid indentation matrix and single indentation](image)

### 3.3. Elastic modulus and hardness

According to the load–displacement curve, the mechanical parameters, such as the hardness and Young modulus, can be calculated using Eq. (1)–(5). In this study, the averaging method was employed for data analysis. Table 2 shows the mechanical parameters obtained from the nanoindentation.

The samples parallel to the bedding have an average Young’s modulus of 46.75 GPa and average hardness of 2.57 GPa; the samples perpendicular to the bedding have an average Young’s modulus of 50.09 GPa and average hardness of 2.98 GPa; and the drill cutting samples have an average Young’s modulus of 46.01 GPa and average hardness of 2.51 GPa. It can be concluded that the samples perpendicular to the bedding have the highest Young modulus and hardness, whereas the drill cutting samples have the lowest values. The reason for this is that the drilling fluids affect the shale properties at the nanoscale level. Moreover, the samples parallel to the bedding exhibit slightly lower Young’s modulus than those perpendicular to the bedding. The difference between the responses in the two different directions indicates the anisotropic characteristics of the shale samples. At the nanoscale level, as the indentation points are on different minerals, the tested mechanical parameters will somewhat fluctuate. Therefore, to obtain representative and reliable mechanical parameters, more indentation points need to be tested to mitigate the uncertainty caused by the heterogeneity of the shale.

| Sample                        | Young’s modulus/GPa | Hardness/GPa |
|-------------------------------|---------------------|--------------|
|                               | Minimum     | Maximum     | Average | Minimum | Maximum | Average |
| Parallel to bedding           | 37.86       | 59.63       | 46.75   | 1.71    | 3.83    | 2.57    |
| Perpendicular to bedding      | 39.43       | 62.11       | 50.09   | 1.81    | 4.37    | 2.98    |
| Drill cuttings                | 35.81       | 55.37       | 46.01   | 1.55    | 3.79    | 2.51    |
Figure 5 shows the maps of the mechanical properties of the nanoindentation areas with samples parallel to the bedding, samples perpendicular to the bedding, and drill cutting samples, respectively. From the figure, it can be seen that the shale is composed of different mechanical phases. The points with higher Young’s modulus represent the hard minerals such as feldspar and quartz. Conversely, the points with lower Young’s modulus indicate soft minerals, including clay minerals and organic matter. Moreover, these images demonstrate that the Young’s modulus and the hardness map correlate very well. Usually, the points with higher Young’s modulus compare to the higher hardness values.

Figure 5. Maps of the mechanical properties of samples parallel to the bedding (a), samples perpendicular to the bedding, (b) and drill cutting samples (c)
3.4. Discussion
Compared to the traditional approaches for obtaining the mechanical properties of rocks, nanoindentation provides a more convenient approach to obtain mechanical information at the nanoscale level. However, these mechanical properties only reflect the micromechanical properties of shale. Moreover, these parameters could not be used directly in drilling analysis, such as wellbore stability. Thus, certain methods that can upscale the mechanical properties from the nanoscale to macroscale level should be used for the data obtained from nanoindentation. For example, the Mori–Tanaka method is one of the most popular methods for data upscaling in nanoindentation [14]. To further demonstrate the accuracy of the upscaling model, the uniaxial compression test is conducted on cylinder shale samples to obtain the stress–strain curve and the Young's modulus. As shown in Figure 6, the Young’s moduli of the samples parallel to the bedding, perpendicular to the bedding, and soaked in drilling fluids are 51.67, 57.93, and 43.26 GPa, respectively. The primary difference between the results of the nanoindentation and uniaxial compression tests is that the data obtained from nanoindentation is much scattered, whereas the results of the compression test is single and at the macroscale level.

**Figure 6. The uniaxial compression stress–strain curves of shale samples**

Generally, shale can be considered as a composite as a porous matrix where inclusions, solid individual minerals, and organic matter are randomly distributed. Evidently, the results at the nanoscale level cannot represent macroscopic performance; hence, the upscaling of shale is a challenging work. There may be multiple solutions because of the complexity of the shale’s composition. Moreover, the limitation of the upscaling methods is that the target porous media microstructure is notably simplified. For matrix–inclusion morphologies that contain up to 40% inclusion, the Mori–Tanaka method can provide better results of homogenized bulk modulus than other upscaling methods. However, if the inclusion is >40%, the prediction error of the upscaling results using the Mori–Tanaka method increases [14]. Moreover, these methods are not capable of classifying the mechanical phases in a single mineral level. For example, quartz and feldspar have similar stiffness values, which were classified into the same mechanical phase. Therefore, it is crucial for researchers to compare macroscale measured data with mathematical methods from nanoscale measurements to obtain more accurate results.
Moreover, the sink-in phenomenon can be observed in the shale surface following nanoindentation because the surface and the interior of the shale are composed of both soft materials such as clay or kerogen [20, 24]. However, if the surface is composed of clay or kerogen while the interior of the shale is composed of hard minerals, such as quartz or carbonate minerals, the pile-up event occurs. This squeeze deformation will cause the indentation height to be higher than the surface average height around the contact boundary. Specifically, when pileup is evident, accurately determining the true contact area is difficult; thus, the contact hardness and Young modulus is overestimated. Therefore, to obtain representative and reliable mechanical parameters, additional indentation points are required to be tested to mitigate this phenomenon.

4. Conclusions
In this study, grid nanoindentation was applied to analyze the mechanical properties of Longmaxi Formation Shale in Zigong area, Sichuan Basin. It has been shown that the elastic properties could be obtained by performing grid nanoindentation.

As can be seen from the load–displacement curves, it can be concluded that the sample perpendicular to the bedding has the highest permanent depth of penetration, whereas the drill cutting samples have the lowest values. Failure in indentation indicates that the number of microcracks in the central of the indentation is much less than that in the contact area of the indenter edge. Moreover, samples perpendicular to the bedding have the highest Young modulus and hardness, whereas the drill cutting samples have the lowest values. Furthermore, the samples parallel to the bedding have slightly lower Young modulus than those perpendicular to the bedding. The maps of the mechanical properties demonstrate that the Young modulus and the hardness map correlate very well. The results of this study indicate that the theory of nanoindentation could be applied to study the mechanical properties of shale at the nanoscale level. Future work should focus on the upscaling of the mechanical properties in nanoscale to macro and reservoir scale.

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