Assessment of deep shale fracture toughness using nanoindentation tests

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Abstract: In the past decade, the extraction of shale gas has become economically feasible due to innovations in horizontal drilling and multistage hydraulic fracturing. This has led to increased interest in deeper understanding of the mechanical properties of shale rocks. Currently, the deep Longmaxi shale with a burial depth of more than 3500 m has been receiving increasing interest in the Sichuan Basin. The first task of large scale development of these reservoirs is to understand the mechanical properties of deep shale rocks. Usually, these properties, such as the Young's elastic modulus $E$, hardness $H$, fracture toughness $K_{IC}$ and other parameters are commonly obtained from laboratory tests with core plugs and logging data interpretations. However, these methods have their own limitations. In recent years, the nanoindentation test has been widely used to predict mechanical properties of shale and other rocks. The advantage of this test is the small sample volume required, which means that drilling cuttings or fragments, which are easily obtained during drilling, can be used. In this study, the nanoindentation method is adopted for the accurate measurement of $K_{IC}$, which is of vital importance for the better understanding of the drilling-induced tensile fractures and for the design of hydraulic fracturing. The specimens retrieved from a deep well in the Sichuan Basin are examined using nanoindentation tests to obtain $K_{IC}$. The results show that $K_{IC}$ is strongly dependent on mineralogy and is anisotropic. Usually, $K_{IC}$ values measured parallel to the bedding planes are generally higher than the corresponding values measured normal to the bedding planes. The obtained fracture toughness can be used to predict the fracability and brittleness of shale, which have instructional significance for drilling and hydraulic fracturing.

1. Introduction
With the rapid increase in natural gas demand, the shale gas resources in the Sichuan Basin have been the hottest target plays in the past decade [1-2]. Usually, the main development shale reservoir is the Longmaxi Formations at a depth of less than 3500 m. With exploration and development, it has been found that the deep shale gas with a burial depth more than 3500 m has a wide distribution and good prospects for exploration, such as the Zigong area, Luzhou area, and Rongchang area [3]. To better develop the deep shale gas reservoir, having a good knowledge of the geomechanical properties of shale is very important from the beginning phase of pre-drill well planning and continuing into...
wellbore stability and hydraulic fracturing [4]. The previous research results show that the
gemachanical properties of shale are mainly controlled by the mineralogy and structural properties,
which mean that shale has obviously anisotropic and heterogeneous features.

The gemechanical properties, such as the Young’s modulus (E), hardness (H), fracture toughness
\(K_{IC}\) and so on, are essential to analyze the stability of wellbores and understand the propagation of
fractures induced by hydraulic fracturing and drilling. These properties can be obtained from routine
laboratory experiments. However, such experiments have their own limitations. In unconfined/triaxial
compression tests, the obtaining standard sized core plugs from downhole cores would be challenging
and the cost is very high [4]. In the cracked Brazilian disk test to get the fracture toughness, a notch is
first created in the center of the sample, which is very difficult to achieve with reliable results. So, to
address the difficulty in measuring rock properties, an advanced nanoindentation test is introduced to
obtain the mechanical parameters [5-14]. The nanoindentation test requires only a small volume of
rock sample from drilling cuttings or fragments, which are easily obtained during drilling and often
cover large intervals of the well trajectory. In recent years, this test has been widely used to investigate
the properties of shale at very small scales.

Essentially, nanoindentation is a technique based on a hard tip, which is pressed into the surface of
a sample [4]. This method can reflect the mechanical properties of the samples on the nanoscale. In the
measuring of fracture toughness in shale, Liu (2015) was the first one to carry out this kind of test to
investigate the fracture toughness of Antrim shale and Opalinus clay shale [5]. Later, Liu et al. (2016)
also obtained the fracture toughness of Bakken samples using nanoindentation; the average fracture
toughness was 3.06 MPa/m\(^{0.5}\) [6]. Followed the study of Liu et al. (2016), Zeng et al. (2017, 2019)
pointed out that the fracture toughness calculated by Liu neglected the holding stage and proposed a
equation to obtain more reliable results [7-8]. Su et al. (2019) compared Liu's method and Zeng's
method and the results show that the average \(K_{IC}\) for these two methods are 10.569 MPa/m\(^{0.5}\) (Liu) and
1.523 Mpa/m\(^{0.5}\) (Zeng) [9]. In addition, Yang et al. (2018) measured fracture toughness on shale and
carried out fluid sensitivity studies, which showed that the water-weakening effect causes fracture
toughness reductions of up to 43% [10]. Gupta et al. (2020) also measured the fracture toughness of
Marcellus, Wolfcamp, Woodford, and Eagle Ford shale using nanoindentation. They found that the
fracture toughness is strongly dependent on mineralogy and is anisotropic; on average 33% higher
when measured parallel to the bedding plane compared to measurements normal to the bedding [11].

In this study, the fracture toughness of deep shale collected from Longmaxi in the Zigong area was
tested using nanoindentation. The first part is dedicated to briefly introducing the nanoindentation
methods and the calculation methods of fracture toughness. This is then followed by the data analysis
and discussion of fracture toughness in nanoindentation tests. Finally, some conclusions are obtained.

2. Experimental methods

2.1. Nanoindentation test

The nanoindentation test is based on a hard indenter tip with known properties that is pressed into the
sample surface [12]. The indenter used in nanoindentation is the Berkovich indenter, which has a
three-side symmetric shape. Normally, nanoindentation tests have three stages, which are; loading,
holding, and unloading. Fig.1 (a) shows the indenter for sensing the load and its associated
displacement into the sample. Fig.1 (b) is the typical load-displacement curve in nanoindentation. In
the loading stage, the applied load increases as the penetration depth increases, and this stage includes
the elastic and plastic deformation. Next, the holding stage is followed to uncover the time-dependent
deformation mechanisms of samples. The unloading stage is the elastic recovery process.
The mechanical properties, mainly hardness $H$ and Young’s modulus $E$, can be obtained from the load-displacement curve. Based on the curve, the following parameters are acquired: the maximum load $P_{\text{max}}$, the maximum displacement $h_{\text{max}}$, the permanent depth of penetration $h_c$ and the elastic unloading stiffness $S = dP/dh$, which is quantified as the slope of the upper portion of the unloading curve, given by [5, 6]:

$$S = \frac{dP}{dh} = \beta \frac{2A_c}{\sqrt{\pi}} E_r$$  \hspace{1cm} (1)

Where $\beta$ is a constant dependent on the geometry of the indenter ($\beta = 1.034$ for a Berkovich indenter), $A_c$ is the contact area between the indenter and the sample, $E_r$ is the reduced Young’s modulus, and $E_r$ is given by [5, 6]:

$$\frac{1}{E_r} = \frac{1}{E} + \frac{1-\nu^2}{E_i}$$  \hspace{1cm} (2)

Where $E$ and $\nu$ are the Young’s modulus and Poisson’s ratio of the samples, and $E_i$ and $\nu_i$ are the Young’s modulus and the Poisson’s ratio of the indenter. For a Berkovich indenter, $E_i = 1141 \text{ GPa}$ and $\nu_i = 0.07$.

The Young’s modulus $E$ of a sample can then be determined by the combination of Eq. (1) and Eq. (2). The hardness can be derived by the following equation [5, 6]:

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

Where $P_{\text{max}}$ is maximum load. The contact area $A_c$ is the function of the contact depth $h_c$ is [5, 6]:

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

The relationship between $h_c$ and $h_{\text{max}}$ [5, 6] is:

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

Where $\varepsilon$ is a geometric constant for the indenter ($\varepsilon = 0.75$ for a Berkovich indenter).

### 2.2. Determination of fracture toughness

In the measurement of fracture toughness on shale there are normally two methods; the crack length-based method and the energy method. However, the first method is not applicable to shale. Gupta et al. (2020) found that reliable fracture toughness values could not be estimated using the crack length-
based method on shales [11]. Rather, the energy method has been used to calculate fracture toughness of shale extensively.

In the energy method, the total energy $U_t$ during the whole indentation process is a sum of the elastic $U_e$, pure plastic $U_{pp}$, and fracture $U_{frac}$ energies. To calculate fracture toughness, the fracture energy $U_{frac}$ is the core required parameter. The calculation process is as follows: firstly, the total energy $U_t$ and elastic energy $U_e$ are calculated according to the nanoindentation load-displacement curve [7-11]:

$$U_t = \frac{P_{max}h_{max}}{3}$$  \hspace{1cm} (6)

$$U_e = \frac{P_{max}h_{max}}{3} \left[ 1 - \frac{3}{2} \left( \frac{h_f}{h_{max}} \right)^2 + 2 \left( \frac{h_f}{h_{max}} \right)^3 \right]$$  \hspace{1cm} (7)

Then, the pure plastic energy $U_{pp}$ is obtained [5-6]:

$$U_{pp} = \left[ 1 - \left( \frac{1 + 3}{2} \left( \frac{h_f}{h_{max}} \right)^2 + 2 \left( \frac{h_f}{h_{max}} \right)^3 \right) \right] U_t$$  \hspace{1cm} (8)

Where $h_f$ is the residual displacement. However, Eq.(8) neglects the maximum holding stage, which sees a much high fracture toughness. Zeng et al (2017) proposed a new equation in which the holding step was considered [7]:

$$U_{pp} = \left[ 1 - \left( \frac{1 + n}{1 + m} \left( 1 - \frac{h_f}{h_{max}} \right) \right) \right] U_t$$  \hspace{1cm} (9)

Where $h_l$ is the loading displacement. In fact, the pure plastic energy $U_{pp}$ obtained from the above two equations does not differ by much. The $U_{pp}$ is 0.922 J from Eq.(8) while the $U_{pp}$ is 0.975 J from Eq.(9) [9]. However, the study of Zeng et al (2017) verified that a decrease of $U_{pp}/U_t$ by 20% can induce a significant increase of fracture toughness by 86% for a shale sample [7]. Hence, Eq.(9) is recommended for the calculation of fracture toughness in this paper.

Thirdly, the fracture energy $U_{frac}$ is calculated as follows [10-11]:

$$U_{frac} = U_t - U_e - U_{pp}$$  \hspace{1cm} (10)

Finally, the fracture toughness $K_{IC}$ is defined as [10-11]:

$$K_{IC} = \sqrt{\frac{U_{frac}}{A_c} E_r}$$  \hspace{1cm} (11)

2.3. Experimental samples

The samples were drilled from full-size cores at a depth of 3934.08 m of Longmaxi shale in the Luzhou area, Sichuan Basin. Nanoindentations were performed in two directions; parallel to bedding and normal to bedding. Here, a cubic sample with length of 20 mm is cut from the full-size cores and the surfaces are parallel to bedding and normal to bedding, respectively. The other sample is made from the drilling cuttings that are obtained from the same depth in an adjacent well which is drilled
using oil-based drilling fluids. Fig.2 shows the cubic sample and drilling cuttings sample used in this study.

![Sample images](image)

(a) The sample made from full-size core and the schematic of test direction

(b) The sample made from drilling cuttings

Fig.2 Experimental samples used in nanoindentation tests

The mineral composition analysis shows that the shale sample is mainly composed of quartz minerals, plagioclase, carbonate minerals (calcite, dolomite), and clay minerals. Due to the high fractions of hard minerals, especially quartz at a fraction of 51.14 wt %, it can be inferred that the Longmaxi shale from the Luzhou area can be expected to be relatively brittle with a high elastic modulus and hardness. The total fraction of clay minerals was 15.81 wt % and the dolomite made up 8.88 wt %. The fraction of calcite, which is considered as a cementing or bonding material that fills the natural pores and cracks of shale, was 18.37 wt %. Additionally, a small amount of pyrite (1.85 wt %), a common mineral found in shale, was also present.

A grid nanoindentation method was carried out on the polished surface of each sample. Each grid consisted of 10 × 10 grid sets and 20 μm spacing. The maximum displacement was 6000 nm and the holding time was 15 s during the holding stage.

3. Results and discussions

3.1. Analysis of fracture toughness

Fig.3 illustrates the load-displacement curves for tests normal to bedding, parallel to bedding, and drilling cuttings. All curves contain three stages: loading, holding, and unloading. Compared with the curves obtained from samples cut normal to bedding and parallel to bedding, the curves of drilling cuttings are more scattered. This may be caused by the effect of drilling fluid that may weaken the mechanical properties of cuttings. Moreover, the maximum load of samples cut parallel to bedding is higher than that of normal to bedding and drilling cuttings under the same maximum displacement in the tests. This indicates that samples cut parallel to the bedding have the strongest mechanical properties, followed by normal to bedding, with the drilling cuttings having the lowest strength.

Fig.4 depicts the SEM images of the grid indentation matrix and of a single indentation. As shown in Fig.4 (a), there are some intrinsic pores and microcracks in the samples, which likely affect the failure mode of the indentation. Fig.4 (b) shows that micro-fractures are created along the indenter edge.
By fitting the loading and unloading stages of the load-displacement curves using the power-law functions, the parameters $n$ and $m$ for different experimental conditions can be obtained as summarized in Table 1.

| Sample                  | $n$    | $m$    | $R^2$ |
|-------------------------|--------|--------|-------|
| Normal to bedding       | 1.9373 | 1.7536 | 0.9813|
| Parallel to bedding     | 1.9662 | 1.6814 | 0.9761|
| Drilling cuttings       | 1.7785 | 1.3891 | 0.9904|

Using the parameters in Table 1, the fracture toughness of different experimental tests is calculated from Eq.(11) and these are statistically plotted in Fig.5. It is clear that the fracture toughness parallel to bedding is the highest, followed by normal to bedding, and the cuttings are the lowest. This means that the fracture toughness of Longmaxi shale is anisotropic and the drilling fluids could weaken the fracture toughness during drilling [10, 15]. Usually, shale is a typical multi-layer sedimentary rock, which means that fractures are cutting across several bedding planes is more difficult compared to fractures are propagating within a layer. In addition, the drilling cuttings have been soaked in drilling fluids for several hours or even days in the wellbore. Meanwhile, the drilling cuttings could be impacted by the drilling string and borehole wall in the process of flowing back from borehole bottom to surface. The above influences could induce the propagation of micro-fractures or pore enlargement. This could induce a decrease of fracture toughness in the drilling cutting samples. From the calculation,
it can be concluded that the average fracture toughness of samples cut normal to the bedding and parallel to the bedding, and of drilling cuttings is 5.346 MPa·m$^{0.5}$, 6.103 MPa·m$^{0.5}$ and 4.461 MPa·m$^{0.5}$, respectively.

![Fracture Toughness vs. Young's Modulus](image)

**Fig.5** The fracture toughness of samples cut normal and parallel to the bedding and of drilling cuttings

**Fig.6** shows the correlation between fracture toughness, Young’s modulus, and hardness for samples cut normal and parallel to the bedding, and of drilling cuttings. As shown in the results, there is a weakly linear relationship between fracture toughness and Young’s modulus or hardness. This means that, as the Young’s modulus or hardness increases, the fracture toughness increases linearly. It can be concluded that a higher Young’s modulus or hardness can make the fractures difficult to propagate. In addition, the fracture toughness used in field applications should consider the anisotropy and the weakening effect of drilling fluid.

![Correlation between Fracture Toughness and Young's Modulus](image)

(a) Plots of fracture toughness vs. Young's modulus
3.2. Discussion

Compared to traditional approaches to evaluate shale fracture toughness, the results obtained from nanoindentation provide higher numerical values than the conventional method. Obviously, the results of nanoscale indentation cannot represent macroscopic performance. Xiong et al (2019) measured the fracture toughness of Longmaxi Formation shale based on the cracked chevron notched Brazilian disc method and the average of fracture toughness parallel to bedding and normal to bedding were 0.5614 MPa·m$^{0.5}$ and 0.6981 MPa·m$^{0.5}$, respectively [16]. The difference between the results of nanoindentation and macroscopic experiments shows that shale is a complex sedimentary rock. Determining how to relate the mechanical properties of the rocks from the nanoscale to the macroscale properties of shale is a difficult task. The complexity of shale’s composition and its anisotropic properties could cause multiple solutions to incorrectly estimate the rock properties on the large scale [15-18]. The data obtained from nanoindentation should be used to develop a method to upscale the mechanical properties obtained from the nanoscale level up to the macroscale. For example, the Mori-Tanaka method is one of the most popular methods for data upscaling in nanoindentation. In the near future, reasonable relationships between nanoscale and macroscale will be crucial for practical application of the nanoindentation method. To further demonstrate the accuracy of the upscaling model, the results of conventional Brazilian disc tests and nanoindentation need to be further studied and compared.

In addition, Eq.(8) has been extensively used in calculating fracture toughness in nanoindentation. This equation is not considered the maximum holding stage and it has produced highly variable estimations of fracture toughness for Longmaxi shale [14]. This is because this method assumes that shale is an ideal elastic material. Hence, this equation may not be suitable for calculation of the fracture toughness in shale, which also shows certain plastic properties and has been verified in previous research.

4. Conclusions

This study measures fracture toughness of deep Longmaxi shale obtained from the Zigong area using grid nanoindentation. The energy-based method is used to calculate the fracture toughness and the results can be summarized as follows:

1) The fracture toughness is strongly anisotropic and can be weakened by drilling fluids. The average fracture toughness normal to the bedding and parallel to the bedding are 5.346 MPa·m$^{0.5}$ and

![Fracture Toughness vs. Hardness](image)

(b) Plots of fracture toughness vs. hardness

Fig.6 Plots of fracture toughness vs. (a) Young's modulus and (b) hardness
6.103 MPa·m$^{0.5}$, respectively. Meanwhile, the average fracture toughness of drilling cuttings is 4.461 MPa·m$^{0.5}$, which is much lower than that of the samples obtained from full-size cores.

2) There is a weakly linear relationship between fracture toughness and Young’s modulus or hardness. Higher Young’s modulus or hardness can make the fractures difficult to propagate.

3) The nanoindentation method provides higher fracture toughness than the conventional macroscopic experiments. The method used to calculate the fracture toughness should consider the maximum holding stage to get a reasonable result. Finally, determination of a reasonable relationship between the nanoscale and macroscale properties will be crucial for practical applications and should be the subject of future work.

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