Application of nanoindentation creep tests to characterize viscoelasticity behavior of deep Longmaxi shales

Lichun Jia, Jing Bai, Jiazhen Zhang

CCDC Drilling & Production Technology Research Institute
No.88 Zhongshan Road, Guanghan, Sichuan Province, China

jialcsc@cnpc.com.cn; ORCID: 0000-0002-2230-0333

Abstract: The development of deep shale gas reservoirs has become economically and commercially viable through novel horizontal well completion and multistage hydraulic fracturing technologies in recent years. In the Sichuan Basin, the deep Longmaxi shale is a major target formation that has attracted increasing interest. Shale is generally characterized as a sedimentary rock with fine grains, high clay, and organic content, low permeability and porosity, and high heterogeneity and anisotropy. Geomechanical characterization of these organic-rich rocks usually includes an estimation of the elastic modulus, strength properties, hysteresis behavior, and creep deformation. Understanding and characterizing the viscoelastic behaviors of shale is essential because they affect the elastic and fracture properties of rock and cause a series of drilling problems related to the wellbore stability, such as borehole shrinkage, pipe sticking, and casing collapse. The viscoelastic behavior of specimens retrieved from a deep well in the Sichuan Basin was examined using nanoindentation creep tests. 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 small sample volume, such as drilling cuttings or fragments, which are obtained easily during drilling. The results indicated that the creep modulus parallel to bedding and normal to bedding is similar, while the creep modulus of drilling cuttings is the lowest. A linear relationship was both observed between the creep modulus and hardness, Young’s modulus for deep Longmaxi shale. The Burgers creep model showed good agreement with the experimental data. The creep behavior of Longmaxi shale exhibited anisotropy, and the effect of drilling fluids could aggravate the creep displacement. These results have elucidated the creep properties in engineering problems of shale gas formation, drilling, and stimulation.

1. Introduction

Deep Longmaxi shale at a depth of more than 3500 m has recently become the main target in the Sichuan Basin [1-2]. Typical deep shale reservoirs are found in the Zigong, Luzhou, and Rongchang areas [3]. Having a good knowledge of the geomechanical properties of shale is very important for
developing deep shale gas reservoirs. Shale is a typical elastic and brittle sedimentary rock [4]. However, shale also shows viscoelasticity behavior or creep properties because of the clay and organic matter contents [5]. The time-dependent behavior of shale could cause the closure of fractures and a decrease in conductivity during the shale gas production period [6-8]. Meanwhile, the creep properties also induce wellbore instability problems, such as borehole shrinkage, pipe sticking, and casing collapse. Therefore, an examination of the viscoelasticity behavior of shale has been the focus over the last decade.

The creep properties of rock are generally investigated using uniaxial creep tests and multistage triaxial creep tests using core samples [6-9]. Numerous experimental and theoretical studies have been performed to examine the creep properties of shale. Li and Ghassemi (2012) reported that the relationship between the creep strain magnitude and time can be analyzed using a power-law function. Moreover, the creep behavior of Barnett, Haynesville, and Marcellus shale is dependent on the shale composition [6]. Sone and Zoback (2014) found that the strain response is proportional to the axial differential load but not to the confining load, and the correlation between the creep strain and time followed a power-law function [7]. Rassouli and Zoback (2018) compared the short-term and long-term creep experiments on shale and carbonates and found that a simple power-law model can describe creep over multiple time intervals [8]. Geng et al. (2018) reported that the differential stress and the bedding orientation could affect the creep behaviors of TourMAKE rare based on a series of triaxial creep experiments [9]. These conventional experiments are very useful for understanding the viscoelasticity behavior of shale. However, these tests are time-consuming and expensive [10]. Moreover, the contribution of individual constituents to the creep behavior and the controlling factors are still poorly understood in the macroscale tests.

The difficulty in measuring creep properties of shale was addressed by introducing a nanoindentation test to analyze the viscoelasticity behavior using a small sample volume [11-12]. Obtaining test samples, such as the drill cuttings and fragments, is very convenient. Nanoindentation is a good technique for measuring the mechanical properties of organic-rich shale on the nanoscale [13-14]. This technique can also quantify the creep behavior of shale [11-12]. Liu et al. (2018) applied a grid nanoindentation method as a function of time to quantify the creep behavior of shale rock. They concluded that shale samples become softer and more prone to fracture growth as the creep time increases [15-16]. Liu et al. (2020) studied the creep behavior using the triaxial and nanoindentation creep tests. Their results showed some discrepancies between the contact creep modulus from the triaxial test and the homogenized contact creep modulus from nanoindentation experiments [17]. Sharma et al. (2019) investigated the effect of temperature on creep properties of organic-rich shales by nanoindentation over a range of temperatures (23°C–350°C). The results revealed a decrease in the creep rate at 300°C for both parallel and perpendicular to the bedding plane [18]. Shi et al. (2020) examined the viscoelastic behavior of shale by nanoindentation creep tests with different holding times. They found that the creep rate decreases rapidly with increasing time at the primary stage. Moreover, the increase in the creep time decreased the mechanical strength properties. In addition, three creep models were used to fit the experimental results [19]. Nevertheless, the viscoelasticity behavior of deep Longmaxi shales has not been studied yet.

In this study, the creep properties of deep shale were tested by nanoindentation creep tests. First, the creep modulus was calculated. The creep displacement-creep time curve of shale was fitted using the Burgers creep model. The creep mechanism was also examined. This study is meaningful for understanding the intrinsic creep mechanism of shale on the nanoscale.

2. Methodology and experiments

2.1. Nanoindentation test

The principle of the nanoindentation test is that a hard indenter tip is pressed into the sample surface under a constant loading rate or constant strain rate [13-14]. Normally, there are three stages during testing: loading, holding, and unloading. Fig.1 (a) shows the indenter for sensing the load and its
associated displacement into the sample. Fig.1 (b) presents a typical load-displacement curve in nanoindentation. The mechanical properties, hardness, and Young’s modulus can be obtained from the load-displacement curve. Besides, the viscoelasticity behavior of the sample can be characterized by examining the experimental results in the holding stage.

Fig.1 Schematic illustration of nanoindentation and load-displacement curve

The hardness can be derived using the following equation [13]:

$$H = \frac{P_{\text{max}}}{A_c},$$

where $P_{\text{max}}$ is the maximum load. The contact area $A_c$ is a function of the contact depth $h_c$ [13]:

$$A_c = 24.5h_c^2,$$

A relationship was observed between $h_c$ and $h_{\text{max}}$ [13]:

$$h_c = h_{\text{max}} - \epsilon \frac{P_{\text{max}}}{S},$$

where $\epsilon$ is a geometric constant for the indenter ($\epsilon = 0.75$ for a Berkovich indenter). The Young’s modulus of the sample can then be determined:

$$\frac{1}{E_r} = \frac{1 - \nu^2}{E} + \frac{1 - \nu_i^2}{E_i},$$

where $E$ is the Young’s modulus, $E_r$ is the reduced Young’s modulus, $\nu$ is the 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, $E_i = 1141$ GPa and $\nu_i = 0.07$.

2.2. Creep modulus and model

The contact creep modulus $C$ is generally used to characterize the nanoindentation creep behavior. A greater contact creep modulus of a material indicates a lower creep rate. The calculation of the creep modulus was obtained using the compliance method [20].

The creep compliance rate $\dot{L}(t)$ of the nanoindentation holding stage can be obtained using the following equation [12, 17-18]:

$$\dot{L}(t) = \frac{2a_U \dot{h}(t)}{P_{\text{max}}},$$

where $\dot{h}(t)$ is the rate of the indentation depth; $P_{\text{max}}$ is maximum load throughout the test; and $a_U$ is the radius of the contact between the indenter and sample surface upon unloading, which is obtained using $a_U = \sqrt{A_c/\pi}$ (Slim et al., 2018).
The contact creep compliance $L(t)$ can be obtained by integrating Eq. (5) with respect to time from the beginning of the holding stage [17-18]:

$$L(t) - L(0) = \frac{2a_i \Delta h(t)}{P_{\text{max}}} = \frac{\ln \left( \frac{t}{\tau} + 1 \right)}{C}$$  \hspace{1cm} (6)

where $h(t)$ is the change in the indentation depth; $C$ is the contact creep modulus; and $\tau$ is the characteristic viscous time. Vandamme et al. (2012, 2013) reported that the contact creep compliance $L(t)$ is a material property that conveys as much information about the viscoelastic response of the material as the more common uniaxial creep compliance. The contact creep modulus $C$ has the same dimensions as a modulus, and the greater the $C$, the lower is the creep rate of the material. The contact creep modulus $C$ can be calculated using the following equation [17, 21]:

$$C = \frac{P_{\text{max}}}{2a_i x_1}$$  \hspace{1cm} (7)

where $x_1$ is the parameter derived by fitting the holding stage depth $\Delta h$ and time $t$ with $\Delta h(t) = x_1 \log t + \text{const}$.

Shi et al. (2020) examined the viscoelasticity of shale in nanoindentation using three different rheological models: the three-element Voigt, Burgers, and two-dashpot Kelvin model. The results indicated that the three models could capture the time-dependent behavior of shale. However, the standard deviation of the two-dashpot Kelvin model was the highest, indicating that more fitting parameters can induce model instability [19]. Liu et al. (2018) used the Burgers model to characterize the creep curve of the shale with good fitting results. Therefore, in this study, the Burgers model was used to fit the creep behavior [15]. The Burgers model combines the Maxwell and Kelvin-Voigt model, which is suitable for defining the first and the second stages of the creep curve. The equation of the Burgers creep model is given by the following:

$$h^2(t) = \frac{\pi}{2} P_{\text{max}} \cot \alpha \left(1 - \nu^2\right) \left[ \frac{1}{E_1} + \frac{1}{E_2} (1 - e^{-\lambda t}) + \frac{1}{\eta(1 - \nu^2)} t \right]$$  \hspace{1cm} (8)

where $E_1$ and $E_2$ are the moduli and $\lambda$ is creep time constant; $\eta$ is the long-term creep viscosity (GPa s); and $\nu$ is the Poisson’s ratio.

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

(a) Sample made from a full-size core and a schematic of the test direction
Mineral composition analysis showed that the shale sample was composed mainly of quartz minerals, plagioclase, carbonate minerals (calcite and dolomite), and clay minerals. The high fractions of hard minerals, particularly quartz, at a fraction of 51.14 wt %, suggest that the Longmaxi shale of the Luzhou area is expectedly relatively brittle with a high elastic modulus and hardness. The total fraction of clay minerals and dolomite was 15.81 wt % and 8.88 wt %, respectively. The fraction of calcite was 18.37 wt %, which is considered as a cementing or bonding material that fills the natural pores and cracks of shale. A small amount of pyrite (1.85 wt %), a common mineral found in shale, was also present.

A grid nanoindentation method was conducted on the polished surface of each sample. Each grid was a $5 \times 5$ grid set with a 20 μm spacing between each indentation. In addition, as the holding time plays a major role in the creep properties of shale, different holding times of 15 s, 60 s, and 200 s at the holding stage were used in the test.

3. Results and discussion

3.1. Analysis of the creep modulus
According to Eq.(7), the creep modulus was calculated at a holding time of 200 s. Fig.3 illustrates the creep modulus for nanoindentation parallel to bedding, normal to bedding, and drilling cuttings. The creep modulus parallel to bedding and normal to bedding is roughly equal, while the creep modulus of drilling cuttings is the lowest. This means that the drilling fluids could weaken the mechanical properties of shale during drilling, and the in situ shale shows more obvious creep behavior. The mean values of results parallel to bedding, normal to bedding, and drilling cuttings were 976.9 GPa, 996.3 GPa, and 824.4 GPa, respectively.
Fig. 4 shows a linear relationship between the creep modulus and hardness, Young’s modulus for deep Longmaxi shale. Overall, the contact creep modulus increased with increasing hardness or Young’s modulus.

![Graphs showing correlations between creep modulus (C) and hardness (H), and between C and Young’s modulus (E).](image)

(a) Correlations between C and H  
(b) Correlations between C and E

Fig.4 Correlations between the creep modulus C and (a) hardness H and (b) Young’s modulus E

3.2. Creep behavior modeling

The Burgers creep model was used to fit the experimental displacement-time curves of the holding stage. Fig.5 shows the curves of experimental data and fitting data for the results parallel to bedding, normal to bedding, and drilling cuttings. As the holding time was increased, the displacement rapidly increased before 75 s and gradually became steady. A comparison of the displacement with different holding times of 15 s, 60 s, and 200 s showed that the complete creep behavior of shale would not be determined at a short holding time. For creep modeling, the Burgers creep model indicated good agreement with the experimental data. For different tests results, the displacement obtained from the drilling cuttings was much lower than the displacement parallel to bedding and normal to bedding. Conversely, the displacement obtained from normal to bedding was slightly higher than that of parallel to bedding. These results indicated two characteristics of shale: 1) the creep behavior of Longmaxi shale shows anisotropy, and 2) the effect of drilling fluids can aggravate the creep displacement significantly. The reason is that the presence of drilling fluids can reduce the friction between the grains, which could enhance creep deformation.
3.3. Creep mechanism and discussion
Shale is a fine-grained sedimentary rock consisting of a mixture of minerals and organic matter. This complex mineral composition is highly heterogeneous on the nanoscale. Usually, these mineral compositions are divided into different categories: a soft phase representing clay minerals, a hard phase representing quartz, and a medium phase representing other minerals. Each of these components show different creep behavior. It is indisputable that harder the minerals, smaller is the creep deformation. In Liu’s study (2018), a longer creep time indicated a lower volume percentage of hard mineral phase and a higher volume percentage of soft mineral phase. This is because the dislocation of creep can cause dynamic recrystallization and reduce the grain size. Another reason is that when the indenter acts on hard minerals, the soft minerals under the hard minerals will be squeezed [17]. In contrast, Sharma et al. (2019) found that the organic matter decreases the frictional coefficient between the clay particles and organic matter, causing a lower creep rate of the porous clay and kerogen phase than the clay particles [22].

The linear relationship between the creep modulus and hardness and Young’s modulus and the creep behavior anisotropy suggests that the creep mechanism of shale is the particle rearrangement and compaction within the clay composite phase [18]. The drilling cuttings show that the presence of drilling fluids facilitates grain compaction and rearrangement and escalates creep deformation. This is because the drilling fluids can enter the pores and matrix among the particles, decreasing the frictional coefficient between two clay particles and between the clay particles and organic matter [15, 18]. The grain motion by frictional sliding can enhance creep deformation.

The previous studies reported that the minutes-long nanoindentation creep tests could qualitatively capture the long-term creep kinetics of organic-rich shale. The observed trend was qualitatively identical to the trends observed in macroscopic triaxial creep experiments. However, the parameters obtained from nanoindentation, mainly the contact creep modulus, were higher than the contact creep modulus from the triaxial test. Therefore, a reasonable upscaling model between the nanoindentation creep tests and triaxial creep experiments is needed.

4. Conclusions
A nanoindentation method was applied to analyze the creep properties of deep Longmaxi shale. From the results, the following conclusions were derived:

(1) The creep moduli parallel to bedding and normal to bedding are similar, while the creep modulus of drilling cuttings is the lowest. The mean value of results parallel to bedding, normal to bedding, and drilling cuttings is 976.9 GPa, 996.3 GPa, and 824.4 GPa, respectively.
(2) A linear relationship was observed between the creep modulus and hardness, Young’s modulus for deep Longmaxi shale.

(3) The Burgers creep model showed good agreement with the experimental data; the displacement rapidly increased and gradually became steady as the holding time was increased.

(4) The creep behavior of Longmaxi shale showed anisotropy, and the effect of drilling fluids could aggravate the creep process significantly.

References
[1] Zou Caineng, Zhao Qun, Cong Lianzhu, et al. (2021). Development progress, potential and prospect of shale gas in China. Natural Gas Industry, 41(01): 1-14.
[2] Zhang Jinchuan, Tao Jia, Li Zhen, et al. (2021). Prospect of deep shale gas resources in China. Natural Gas Industry, 41(01): 15-28.
[3] Yang Yueming, Chen Yulong, Liu Shenyang, et al. (2021). Status, potential and prospect of shale gas exploration and development in the Sichuan Basin and its periphery. Natural Gas Industry, 41(01): 42-58.
[4] Hiroki Sone, Mark D. Zoback. (2013). Mechanical properties of shale-gas reservoir rocks - Part 1: Static and dynamic elastic properties and anisotropy. Geophysics, Vol.78, No.5, 381-392.
[5] Hiroki Sone, Mark D. Zoback. (2013). Mechanical properties of shale-gas reservoir rocks - Part 2: Ductile creep, brittle strength, and their relation to the elastic modulus. Geophysics, Vol.78, No.5, 393-402.
[6] Li Y, Ghassemi A (2012). Creep behavior of Barnett, Haynesville, and Marcellus shale. In: 46th US rock mechanics/geomechanics symposium. Am Rock Mech Assoc Chicago, Illinois, 24-27.
[7] Hiroki Sone, Mark D. Zoback. (2014). Time-dependent deformation of shale gas reservoir rocks and its long-term effect on the in situ state of stress. International Journal of Rock Mechanics & Mining Sciences, 69: 120-132.
[8] Fatemeh S. Rassouli, Mark D. Zoback. (2018). Comparison of Short-Term and Long-Term Creep Experiments in Shales and Carbonates from Unconventional Gas Reservoirs. Rock Mechanics and Rock Engineering, https://doi.org/10.1007/s00603-018-1444-y.
[9] Zhi Geng, Audrey Bonnelye, Mian Chen, Yan Jin, Pierre Dick, Christian David, Xin Fang, Alexandre Schubne. (2018). Time and temperature dependent creep in Tournemire Shale. Journal of Geophysical Research: Solid Earth, 123(11): 9658-9675.
[10] Zhaoyang Ma, Ranjit Pathegama Gamage, Chengpeng Zhang. (2020). Application of nanoindentation technology in rocks: a review. Geomech. Geophys. Geo-energ. Geo-resour, 6:60. https://doi.org/10.1007/s40948-020-00178-6.
[11] Mighani S, Taneja S, Sondergeld CH, Rai CS (2015) Nanoindentation creep measurements on shale. In: 49th US Rock Mechanics/Geomechanics Symposium. Am Rock Mech Assoc 28 June-1 July, San Francisco, California.
[12] Mighani, S., Bernabé, Y., Boulenouar, A., Mok, U., & Evans, B. (2019). Creep deformation in Vaca Muerta shale from nanoindentation to triaxial experiments. Journal of Geophysical Research: Solid Earth, 124 (8): 7842-7868.
[13] Liu, K., Ostadhassan, M., Bubach, B. (2016). Applications of nanoindentation methods to estimate nanoscale mechanical properties of shale reservoir rocks. J. Nat. Gas. Sci. Eng. 35: 1310-1319.
[14] Liu, K., Ostadhassan, M., Bubach, et al. (2018). Statistical grid nanoindentation analysis to estimate macro-mechanical properties of the Bakken Shale. J. Nat. Gas. Sci. Eng. 53: 181-190.
[15] Kouqi Liu, Mehdi Ostadhassan, Bailey Bubach. (2018). Application of nanoindentation to characterize creep behavior of oil shales. Journal of Petroleum Science and Engineering, 167: 729-736.
[16] Kouqi Liu, Mehdi Ostadhassan, Bailey Bubach, Robert Dietrich, and Vamegh Rassouli. (2018). Nano-dynamic mechanical analysis (nano-DMA) of creep behavior of shales: Bakken case study. J. Mater. Sci., 53: 4417-4432.
Kouqi Liu, Fatemeh S. Rassouli, Bo Liu, Mehdi Ostadhassan. (2020). Creep Behavior of Shale: Nanoindentation vs. Triaxial Creep Tests. Rock Mechanics and Rock Engineering, https://doi.org/10.1007/s00603-020-02255-4.

Prashant Sharma, Ravi Prakash, Sara Abedi. (2019). Effect of temperature on nano- and microscale creep properties of organic-rich shales. Journal of Petroleum Science and Engineering 175: 375-388.

Xian Shi, Shu Jiang, Liu Yang, Mingming Tang, Dianshi Xiao. (2020). Modeling the viscoelasticity of shale by nanoindentation creep tests. International Journal of Rock Mechanics & Mining Sciences 127, 104210.

Vernik, L., Milovac, J. (2011). Rock physics of organic shales. Leading Edge, 30 (3): 318-323.

Jianting Du, Andrew J. Whittle, Liming Hu, Thibaut Divoux, Jay N. Meegoda. (2020). Characterization of meso-scale mechanical properties of Longmaxi shale using grid microindentation experiments. Journal of Rock Mechanics and Geotechnical Engineering, https://doi.org/10.1016/j.jrmge.2020.09.009.

Slim, M., Abedi, S., Bryndzia, T.L., Ulm, F.J. (2018). The role of organic matter on nanoand micro-scale creep properties of source rocks. J. Eng. Mech. 145 (1), 04018121.