of constituents in mudstone

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Abstract In deep underground engineering, the creep behaviors of soft rocks have been widely investigated to help understand the mechanism of the time-dependent large deformation and failure of underground engineering structures. However, rocks were used to be regarded as homogeneous materials and there are limited studying results about the time-dependent properties of constituents in them to reveal their creep mechanism. In this context, the targeting nanoindentation technique (TNIT) was adopted to investigate the viscoelastic characteristics of kaolinite and quartz in a two-constituent mudstone sample. The TNIT consists of identifications of mineralogical constituents in mudstone and nanoindentation experiments on identified constituents. After conducting experiments, the unloading stages of the typical indentation curves were analyzed to calculate the hardness and elastic modulus of constituents in mudstone. And the 180 s load-holding stages with the maximum load of 50 mN were transformed to the typical creep strain-time curves for fitting analysis by using the Kelvin model, the standard viscoelastic model and the extended viscoelastic model. Fitting results show that the standard viscoelastic model can perfectly express the nanoindentation creep behaviors of both kaolinite and quartz and fitting constants are suitable to be used to calculate their creep parameters. The creep parameters of kaolinite are much smaller than that of quartz, which drives the time-dependent large deformation of the soft mudstone. At last, the standard viscoelastic model was verified on a sandstone sample.

Keywords Viscoelastic properties · TNIT · Mudstone · Constituents · Creep constitutive model

1 Introduction

Creep property, a time-dependent behaviour, is the inherent attributes of rocks, soft mudstone in particular (J. Sun, 2007). In deep underground engineering, the creep deformations of rocks become more common due to the high stress (He et al., 2005). The time-dependent deformations of rocks negatively affect the mining safety (X. Li et al., 2017; Mishra and Verma, 2015; Sainoki and Mitri, 2017; Zhou et al., 2019), backfill mining (Z. Guo et al., 2019; Q. Sun et al., 2013), coalbed methane production (Danesh et al., 2016; Danesh et al., 2017; Z. H. Guo et al., 2018; Kang et al., 2015), and geo-sequestration (Z. B. Liu et al., 2015; Sone and Zoback, 2014). Consequently, to enhance the understanding of this phenomenon is of importance to guarantee the stability of underground engineering. And macro-scale laboratory creep experimental methods, such as uniaxial and triaxial compression tests, have been broadly used to investigate the rheological deformations of rocks, which threw light on their creep characteristics (Sha et al., 2018; Xiong et al., 2019; S. Q. Yang, Xu, et al., 2015). However, these experiments were too complicated. On the one hand, the load-holding time of some triaxial compressive tests was about several days (Sone and Zoback, 2014; Xu et al., 2014) and that for uniaxial compression tests was above 6 hours (Jia et al., 2018; Y. Yang, Xing, et al., 2015). On the other hand, the specimens used in macro-scale laboratory creep tests needed to meet strict size requirements: the typical cylindrical specimen size was 100 mm×φ50 mm (Mishra and Verma, 2015) and the size of the smaller prismatic samples was 25 mm×25 mm×50 mm (Lu and Wang, 2017). In addition, centimetre-scale laboratory experiments regard rocks consisting of various constituents (Guery et al., 2008) as homogeneous bodies, where the mechanism of large creep deformation of soft rock was not revealed. Consequently, adopting a new technique to identify various mineralogical constituents and investigate their creep characteristics is very meaningful.

Nanoindentation technique, also called the depth-sensing indentation, was first proposed and used by Kalei in 1968 in Russia (Kalei, 1968). This technique has proven to be an effective and convenient method for determining the elasto-plastic mechanical properties of solids based on a small rock sample, most notably elastic modulus, hardness (Oliver and Pharr, 1992) and fracture toughness (Zeng et al., 2019). Merged with CT (Y. Zhang, Lebedev, Al-Yaseri and Yu, 2018; Y. Zhang, Lebedev, Al-Yaseri, Yu, et al., 2018), SEM-EDS (Slim et al., 2019; C. Sun, Li, Gomah, et al., 2020), XRD (Zhao et al., 2019) and 3D...
printing (Kong et al., 2019) observing the microstructures of rock samples, nanoindentation technique was useful to investigate the mechanical behaviors of mineralogical components of shale, granite, coal or mudstone samples. The influence of clay minerals and kerogen on both the hardness and elastic modulus of organic-rich and clay-bearing shale was investigated (Abedi et al., 2016; Alstad et al., 2015). Zhang Fan et al. obtained the hardness and elastic modulus of compositions in granite (Fan et al., 2017). Taking Chinese sub-bituminous coal as a sample, the relationship between its morphologies and its nano-mechanical properties was investigated (Y. Zhang, Lebedev, Al-Yaseri, Yu et al., 2018); Meng et al. and Sun et al. calculated the mean values of the nanoindentation mechanical parameters to represent that of standard coal samples (Meng et al., 2020; C. Sun, Li, Zhang, et al., 2020). For mudstone samples, the nanoindentation technique can test the elastic and plastic mechanics of its minerals (Magnenet et al., 2011) and reveal its heterogeneity (Auvray et al., 2017; F. Zhang, Guo, et al., 2018). However, all geomaterials also exhibit a viscoelastic response, which has improved utilization of this technique on capturing and modelling the creep characteristics of geomaterials. Slim et al. covered the role of the organic matter on creep rates of gas shale by using nanoindentation technique and obtained that organic matter drives its creep rates (Slim et al., 2019). Liu et al. used nanoindentation technique to examine the creep behaviour of multi-phase oil shale by applying Burgers models and the logarithmic function to quantify creep curves of various phases in oil shale (K. Liu et al., 2018; K. Liu et al., 2017). Nevertheless, creep was usually observed as an increase in depth during the load-holding period at maximum load (Slim et al., 2019); and there is no work on how the creep strain-time data can be used to model the nanoindentation creep response. Besides, widely used grid nanoindentation method (K. Liu et al., 2018; K. Liu et al., 2017) complicate the data analysis.

In this context, the nanoindentation technique coupled with XRD, SEM and EDS were used to inspect the creep behaviours of mineralogical constituents in mudstone. This paper is structured as follows: The studied two-constituent mudstone was first presented together with the nanoindentation experimental protocol and experimental method to illustrate the heterogeneous properties of mudstone and the necessity of the targeting nanoindentation technique (Section 2). A theoretical analysis of nanoindentation creep constitutive models and the Oliver-Pharr method followed; the creep models are some viscoelastic models used to fit the nanoindentation strain-time curves (Section 3). The calculating and fitting results of nanoindentation experimental data were then given to distinguish the mechanical properties of two constituents in mudstone and to determine and verify suitable nanoindentation creep model (Section 4).

2 Material and methodology

2.1 Material

A broken soft mudstone block obtained from a deep underground coal mine was researched in this study. As shown in Fig. 1(a), the coal mine named Tongting is located in Anhui province, China; and the sampling layer is above the 7# coal strata deposited at the Permian layer, where the stratigraphic column is presented in Fig. 1(b).

To quantify the mineralogical constituents of this mudstone sample, the XRD experiment was carried out based on the standards No. SY/T 5163-2018; D8 ADVANCE (Bruker, Germany) was used to conduct this test at the range of 2θ (2-θ) from 10° to 65° with the scanning speed of 2°/min. The powder diffraction spectrum is given in Fig. 2(a), which shows that the mudstone consisting of 62.57% kaolinite and 37.43% quartz is a two-constituent geomaterials.

Then, the images of the polished surface of the sample got from SEM-EDS experiments are also given in Fig. 2(b) to observe the distribution of both constituents in the mudstone sample. The left-top one is SEM image,
where there are both smooth and unsmooth surfaces; the smooth surfaces are particle-shaped and embedded in the rest unsmooth surfaces regarded as matrix. The area of unsmooth surfaces is much larger than that of smooth particles, which may represent kaolinite in mudstone and the particles could be quartz. To verify this hypothesis, the right-top EDS image shows the distribution of kalium, silicon, aluminium and oxygen and the left-bottom and right-bottom images give the distributions of a single atom of silicon and aluminium, respectively. Since the chemical equation of kaolinite is \( \text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4 \), and that of quartz is \( \text{SiO}_2 \), the distributions of atoms can help determine the types of minerals in mudstone. The smooth surfaces are the enrichment areas of silicon atom and oxygen atom, and its enrichment degree of silicon atom is much higher than the rest areas. Also, the region does not contain aluminium atoms. EDS images verify that on the surface of this mudstone sample, the smooth particles are quartz and the unsmooth surfaces are kaolinite. The mudstone sample is a two-constituent material having kaolinite as matrix and quartz embedded in it.

![Powder diffraction spectrum](image1.png)

(a) Powder diffraction spectrum to quantify the mineralogical constituents

![Images of SEM and EDS](image2.png)

(b) Images of SEM and EDS for illustrating the distribution of mineralogical constituents

Fig. 2. Diffraction spectrum and images of SEM and EDS.

The mudstone is a typical heterogeneous material composed of clay minerals (kaolinite) and other silt inclusions (quartz). The mechanical properties of it are determined by its various constituents (G. Li et al., 2019), which encourage us to investigate the mechanical performances of its constituents at the meso-scale. Besides, at the beginning of creep experiments, some standard cylindrical samples (100 mm×\( \Phi \)50 mm) were prepared to research the mechanical properties of soft mudstone. However, this experimental plan was dropped due to the influence of macroscale cracks and pores in the mudstone block on the obtaining of reliable mechanical parameters. And a new experimental method, nanoindentation technique, based on small intact samples was used in this study.

2.2 Methodology

2.2.1 Nanoindentation creep experimental protocol
All nanoindentation creep experiments were conducted using a TTX-NHT3 nano-indentor system equipped with a Berkovich tip. The thermal drift of it is less than 0.01 nm/s after allowing thermal equilibrium to be reached for more than 1 h at room temperature (21 °C), which was ignored in this study. As shown in Fig. 3(a), a nanoindentation load-time curve is composed of the loading stage(o-a), load-holding stage(a-b) and unloading stage(b-c), where the 180 s load-holding stage at peak load of 50 mN will be used to analyse creep characteristics (K. Liu et al., 2018). And both the loading and unloading rates are 100 mN/min.

![Fig. 3. Nanoindentation creep experimental protocol illustrated by the nanoindentation load-time curve and nanoindentation creep deformation-time curve.](image)

After nanoindentation test, there is a nanoindentation creep deformation-time curve (Fig. 3b). During the load-holding period, the indenter is displaced further into mudstone. This curve illustrates the portion of displacement that was creep controlled and will be used to analyse the creep characteristics of constituents in mudstone. However, this curve reflects the relationship between creep deformation and time, not that of the typical creep strain and time.

2.2.2 The targeting nanoindentation technique

To make indentations cover representative areas of the material surface and obtain the mechanical properties of all constituents in materials (Slim et al., 2019), the grid nanoindentation method with hundreds of indentations was generally adopted to conduct statistical analysis (K. Liu et al., 2018). However, mechanical parameters obtained from the grid nanoindentation method hinge on the selection of indented surface and the amount of indentations. This experimental method was not of high-efficiency and accuracy. For example, the junction area between mineralogical constituents would affect the accuracies of experimental results.

The whole experimental process to replace the grid nanoindentation method named the targeting nanoindentation technique (TNIT) is graphically illustrated in Fig. 4. A intact cubic mudstone sample from a standardized cylindrical sample were cut into 1.0 cm thick pieces. Experimental surface fixed with epoxy resin was then polished and exposed. The polishing protocol detailed in (C. Sun, Li, Zhang, et al., 2020) was followed. This protocol includes a coarse polishing on 3000 grit emery papers and a fine polishing using oil-based diamond suspension with abrasive grain sizes ranging between 3 and 0.5 μm. SEM image shows that there are many meso-scale cracks on the polishing surface. Thus, to measure their creep properties of both constituents in mudstone and avoid the influence of cracks, the intact surfaces including smooth quartz and unsmooth kaolinite were chosen for probing indenter. Images of EDS and SEM illustrate the types and contents atoms to identify the types of mineralogical constituents in the meso-scale zone. Finally, nanoindentation experiments will be conducted on the both constituents in a targeting way and the mechanical properties of the both constituents in mudstone can be obtained.
The TNIT is a coupled experimental method, where the nanoindentation method is coupled with other microstructure/constituent characterization technique. The constituents of mudstone are required to be observed and identified before conducting nanoindentation experiments. During the observing process, not only the mineralogical constituents are determined, but also the scale separation ($h_{\text{max}} < D/10$, where $h_{\text{max}}$ is the maximum depth of indenter and $D$ is the size of quartz) is satisfied. The technique can be readily incorporated into an inner computer program of nanoindentation system to improve the experimental efficiency on heterogeneous materials.

3 Theories

3.1 Nanoindentation creep model

3.1.1 Nanoindentation strain-time curve

For nanoindentation creep experiments, the stress and strain rate can be written as (Hackney et al., 2012; M.J. and W.D., 1988):

$$\sigma = \frac{\beta F}{h^2(t)}$$  \hspace{1cm} (1)

$$\varepsilon = \frac{dh(t)}{dt} \frac{1}{h(t)}$$  \hspace{1cm} (2)

And then the strain can be obtained by integrating Eq. (2):

$$\varepsilon = \ln \frac{h(t)}{h(0)}$$  \hspace{1cm} (3)

where $\sigma$ is the stress, MPa; $\varepsilon$ is the strain rate, s$^{-1}$; $\varepsilon$ is the strain; $h(t)$ is the indentation depth, nm; $t$ is the creep time, s; $h(0)$ is the indentation depth when creep time is 0, nm; and $\beta$ is an indenter parameter ($\beta=0.0407$ for Berkovich indenter (Hackney et al., 2012)).

Based on the nanoindentation creep data obtained from stage $a$-$b$ in Fig. 3 and Eq. (3), four nanoindentation curves of mudstone are shown in Fig. 5. Two curves labeled 1-1 and 1-2 are got from indentations on kaolinite and other two curves named 2-1 and 2-2 are indented on quartz in mudstone.
Fig. 5. Nanoindentation strain-time curves of kaolinite (1-1 and 1-2) and quartz (2-1 and 2-2) in mudstone.

In Fig. 5 (a), the whole strain-time curves with load-holding time of 180 s are given and their first stages of 15 s are enlarged in Fig. 5 (b) to help observe the initial part of load-holding period. The patterns of nanoindentation strain-time curves of both constituents in mudstone are same: These curves can be divided into (I) transient creep stage and (II) steady creep stage. During the period of the transient creep stage, the strain rates are very large and decrease sharply with time. After recording the steady stage, the strain develops at a constant rate. And the load-holding time of 5 s is the dividing point between two stages. Note that there is no tertiary (accelerating) creep stage and initial instantaneous strain in nanoindentation creep-time curves. Consequently, nanoindentation creep curves can be expressed by viscoelastic creep models that do not include elastic elements in series and plastic elements.

3.1.2 Nanoindentation viscoelastic creep models

Here, based on the patterns of nanoindentation strain-time curves, three nanoindentation viscoelastic creep models were proposed in Fig. 6.
The extended viscoelastic model

Fig. 6. Three nanoindentation creep models: (a) The Kelvin model, with one elastic element and one viscous element in parallel; (b) the standard viscoelastic model, with one viscous element and one Kelvin model in series; and (c) the extended viscoelastic model, with one viscous element and two Kelvin models in series.

For the elastic element in Fig. 6, the stress ($\sigma$) is proportional to the strain ($\varepsilon$), while for the viscous element, it is proportional to the strain rate ($\dot{\varepsilon}$ or $d\varepsilon/dt$). The relationship between stress and strain can be written as Eq. (4) for the elastic element; and that between stress and strain rate for the viscous element are given in Eq. (5).

$$\sigma = E\varepsilon$$  \hspace{1cm} (4)

$$\sigma = \eta\varepsilon \Rightarrow \eta \frac{d\varepsilon}{dt}$$  \hspace{1cm} (5)

where $E$ is the modulus, GPa, and $\eta$ is the viscosity, GPa·s.

According to the stress and strain relationship of the parallel and the series structures in Fig. 6, the creep function of the Kelvin model can be summarized as:

$$\varepsilon = \frac{\sigma}{E} (1 - e^{-\frac{E}{\eta} t})$$  \hspace{1cm} (6)

and, the standard viscoelastic model and the extended viscoelastic model can be collected by:

$$\varepsilon = \sum_{i=1}^{n} \frac{\sigma}{E_i} (1 - e^{-\frac{E_i}{\eta_i} t}) + \frac{\sigma}{E_{n+1}} t$$  \hspace{1cm} (7)

where $n$ is the number of the Kelvin models in series in the creep viscoelastic models. If $n = 1$, Eq. (7) is the standard viscoelastic model; and if $n = 2$, it is the extended viscoelastic model. For fitting the nanoindentation creep strain-time curves, Eq. (7) is further simplified as:

$$\varepsilon = \sum_{i=1}^{n} A_i (1 - e^{B_i t}) + Ct$$  \hspace{1cm} (8)

where $A_i$, $B_i$ and $C$ are fitting constants, and $A_i = \sigma / E_i$, $B_i = E_i / \eta_i$ and $C = \sigma / E_{n+1}$.

3.2 Oliver-Pharr method

The Oliver-Pharr method gives the hardness and elastic modulus of materials by analyzing the unloading stage of the load-depth curve and indentation profile( Oliver and Pharr, 1992, 2004). As shown in Fig. 7(a), a typical load-depth curve can be obtained as the indenter enters into and exists from the surface of mudstone. The curve consists of loading segment (o-a), load-holding segment (a-b), and unloading segment (b-c); also, segment b-d is the curve tangent of unloading segment (c) at its initial portion. In Fig. 7(b), a typical Berkovich indentation is composited by two surface profiles under load (before and after the load-holding segment (b) or creep) and the residual surface profile after load removal.
A load-depth curve from a typical indentation test.

An indentation profile including initial surface, residual surface and surfaces before and after creep deformation.

Fig. 7. Nanoindentation calculating schematic diagram of Oliver-Pharr method.

In Fig. 7, $F_m$ is the peak load exerted by the indenter, mN; $h_m$ is the maximum depth of indentation, nm; $h_c$ is the contact depth of indentation under the peak load $F_m$, nm; $h_k$ is the sinking depth due to load, nm, where $h_c = h_m - h_r$; $h_p$ is the plastic indentation depth after complete unloading, nm; $h_h$ is the creep depth caused by the load-holding segment (b), nm; $h_s$ is the tangent depth of unloading curve, nm; $S$ is the contact stiffness, mN/nm.

It is noted that the load-holding method is mandatory to derive stable unloading data for the calculation of hardness and elastic modulus (Hu and Li, 2015; Shi et al., 2019; Zeng et al., 2019) and the holding time is 180 s in this study (K. Liu et al., 2018). The stable unloading curve is usually well described by a simple power relation:

$$ F = \alpha (h - h_p) \beta, \quad (9) $$

where $\alpha$ and $\beta$ are the power law fitting constants and determined by fitting procedure. Then the contact stiffness is calculated by analytically differentiating this expression at the peak load or the maximum depth, i.e.,

$$ S = \left( \frac{dF}{dh} \right)_{F=F_m,h=h_m} = \alpha \beta (h_m - h_p)^{\beta-1}. \quad (10) $$

The contact depth $h_c$ and the projected area of contact between the indenter and indented constituents $A_c$ are measured by:

$$ h_c = h_m - \varepsilon (h_m - h_r), \quad (11) $$

$$ A_c = 24.5 h_r^2 + C_1 h_r^{1/2} + C_2 h_r^{1/2} + \ldots + C_8 h_r^{1/28}, \quad (12) $$

where $C_1, \ldots, C_8$ are the fitting constants; $\varepsilon$ is the geometric constant for Berkovich indenter ($\varepsilon = 0.75$); $A_c$ is the projected contact area, nm$^2$. Note that the lead term of Eq. (4) describes Berkovich indenter perfectly.

As a result, the hardness is the average load under the indenter tip and reduced modulus can be calculated from BASH formula (Bulychev et al., 1975).

$$ H_{IT} = \frac{F_m}{A_c} \quad (13) $$

$$ E_i = \frac{S \sqrt{\pi}}{2 \eta \sqrt{A_c}} \quad (14) $$
where $H_{IT}$ is the hardness, MPa; $E_r$ is the reduced modulus, GPa; $\eta$ is the geometric constant for Berkovich indenter ($\eta=1.034$). When the effects of non-rigid indenters on the measurement of elastic modulus of mudstone are ignored, reduced modulus can be used to represent it. Otherwise, it can be calculated by

$$\frac{1-v_{IT}^2}{E_{IT}} = \frac{1}{E_r} - \frac{1-v_i^2}{E_i}, \quad (15)$$

where $v_{IT}$ is the Poisson’s ratio of materials and $v_{IT} = 0.3$ in this study; $E_i$ and $v_i$ are the elastic modulus and the Poisson’s ratio of the diamond indenter and $E_i = 1141$ GPa and $v_i = 0.07$.

### 4 Results and discussions

#### 4.1 Hardness and elastic modulus

Two nanoindentation tests were made on two constituents (kaolinite and quartz) in mudstone to calculate their hardness and elastic modulus. In Fig. 8(a) and (b), the nanoindentation load-time curves and indentations on kaolinite are labelled 1-1 and 1-2; and the results of quartz are named 2-1 and 2-2.

![Nanoindentation load-depth curves of kaolinite and quartz](image1.png)

![Indentations on kaolinite and quartz in mudstone](image2.png)

Fig. 8. Load-depth curves of two constituents in mudstone

The load-depth curves on different constituents have the same pattern: During the loading segment, the depths increase sharply with the load at first and then slightly; there is a small creep deformation during the load-holding segment; and after unloading, elastic deformations recover while there are unrecoverable plastic deformations. However, for different constituents, the magnitudes of depths are not the same. The responses associated with the indentations on kaolinite (1-1 and 1-2) give deeper indentation maximum depth (>2000 nm); while, the responses associated with indentation in quartz have lower indentation maximum depths (<700 nm).

Based on the typical Oliver-Pharr method, the hardness and elastic modulus were calculated to distinguish the mineralogical constituents. In Table 1, the mean values of hardness and elastic modulus of kaolinite are 355.75 MPa and 17.79 GPa, respectively. And that of quartz in mudstone are much larger than them, which are 9055.60 MPa and 66.54 GPa. The mechanical performances of kaolinite matrix are much smaller than that of quartz in mudstone, which explains why the argillaceous rocks are soft. For one constituent, the calculated
mechanical parameters are close, meaning the targeting nanoindentation technique can be used to obtain the mechanical parameters of various mineralogical compositions in this mudstone.

**Table 1.** Hardness and elastic modulus of kaolinite and quartz in mudstone.

| No. | E (GPa) | H (MPa) |
|-----|---------|---------|
| 1-1 | 18.76   | 352.51  |
| 1-2 | 16.81   | 358.98  |
| Mean values | 17.79 | 355.75 |
| 2-1 | 67.08   | 8953.40 |
| 2-2 | 66.00   | 9157.80 |
| Mean values | 66.54 | 9055.60 |

4.2 Nanoindentation creep parameters

In this section, the proposed nanoindentation creep viscoelastic models in Fig. 6 will be used to fit the nanoindentation experimental data on kaolinite and quartz in mudstone. Fig. 9(a), (b) and (c) gives the fitting results of Kaolinite by using the Kelvin model, the standard viscoelastic model, and the extended viscoelastic model, respectively. And Fig. 10(a), (b) and (c) shows that of quartz in mudstone.

![Fitting results of kaolinite by using the Kelvin model](image1)

Fitting function of 1-1:
$$\varepsilon = 0.0536(1-e^{-0.0168t})$$

Fitting function of 1-2:
$$\varepsilon = 0.0472(1-e^{-0.0186t})$$

![Fitting results of kaolinite by using the standard viscoelastic model](image2)

Fitting function of 1-1:
$$\varepsilon = 0.0377(1-e^{-0.0111t}+(1.44E-4)t$$

Fitting function of 1-2:
$$\varepsilon = 0.0273(1-e^{-0.0111t}+(1.45E-4)t$$

![Fitting results of kaolinite by using the extended viscoelastic model](image3)

Fitting function of 1-1:
$$\varepsilon = 0.0145(1-e^{-0.0152t})+0.0240(1-e^{-0.0168t})+(1.11E-4)t$$

Fitting function of 1-2:
$$\varepsilon = 0.0134(1-e^{-0.0152t})+0.0168(1-e^{-0.0168t})+(1.24E-4)t$$

**Fig. 9.** Fitting results of kaolinite by using the Kelvin model, the standard viscoelastic model and the extended viscoelastic model.
Fig. 10. Fitting results of quartz by using the Kelvin model, the standard viscoelastic model and the extended viscoelastic model.

Fitting curves by using the Kelvin model did not yield desirable results. As shown in Fig. 9 (a) and Fig. 10 (a), the Kelvin model cannot fit the transient creep stage, where both kaolinite and quartz in mudstone have large strain rates. When describing the final part of the steady creep stage by the Kelvin model, the strain rate is zero, which brings a decidedly contrarian view to the steady creep stage of nanoindentation creep strain-time curves. Consequently, the Kelvin model cannot be used to describe the creep properties of constituents in mudstone. The values of $R^2$ also shows the that the fitting goodness of the Kelvin model is much poorer than the other two viscoelastic models.

For the viscoelastic models consisting of a viscous element and one or several Kelvin models, fitting results are very good. This result shows that the creep model presented by Eq. (8) can be used to describe the nanoindentation creep behaviors of both constituents in mudstone and the more of the number of the Kelvin models, the better of the fitting results. However, with the increase of the number of the Kelvin models, the fitting constants are probably fallacious. For example, as shown in Fig. 10 (c), the coefficient of the first order term of 2-2 is -3.24×10^{-4}, which should be a positive value in reality; and the fitting value of $E_2/\eta_2$ for experiment labeled 2-1 is much larger than that of experiment 2-2. Consequently, for calculating the creep parameters, the standard viscoelastic model is the most useful model among them.

According to the fitting results by using the standard viscoelastic model, the creep parameters of both kaolinite and quartz in mudstone can be obtained, as shown in Table 2.

| No. | $h$/nm | $f$/MPa | $A_1$ | $B_1$/GPa·s | $C\times10^{-4}$/GPa·s | $E_1$/GPa | $\eta_1$/GPa·s | $\eta_2$/GPa·s |
|-----|--------|---------|-------|-------------|----------------|-------|----------|----------|
| 1-1 | 2402.35 | 353.07 | 0.0337 | -0.0874 | 1.44 | 10.48 | 119.87 | 2451.88 |
| 1-2 | 2398.59 | 353.19 | 0.0273 | -0.1254 | 1.45 | 12.94 | 103.17 | 2435.79 |
|     | Mean values |       | 11.71 | 111.52 | 2443.83 |
| 2-1 | 607.66 | 5503.47 | 0.0112 | -0.1723 | 2.06 | 491.38 | 2851.89 | 26715.89 |

Table 2: Creep parameters of kaolinite and quartz in mudstone.
Note that the values of \( h_0 \) was got from the mean values during the load-holding period to minimize the influence of creep deformations on calculated results. And concurring with the above calculated hardness and elastic modulus, the creep parameters of different constituents in mudstone are various, too. For example, the mean value of the first viscous element of kaolinite is 111.52 GPa·s, while this value of quartz is much larger, which is 3066.21 GPa·s. This comparison means that the soft clay minerals in mudstone have large creep deformation, which reveals the mechanism of large creep deformation of argillaceous engineering.

### 4.3 Verification of nanoindentation creep model

To verify that the viscoelastic creep models (Eq. (8)) can perfectly fit the nanoindentation strain-time curves and that the calculated creep parameters may be unreliable when \( n \geq 1 \), four nanoindentation experiments were made on the quartz in a sandstone obtain from Suntuan Coal Mine, Anhui, China. Fig. 11 (a) and (b) give the load-depth curves and indentations of them. The load-depth curves were used to calculate the elastic modulus and hardness and analyse the creep characteristic of quartz in sandstone; the indentations illustrate the application of the targeting nanoindentation technique.

![Quartz](image)

(a) Four load-depth curves of quartz  
(b) Four indentations on quartz

**Fig. 11.** Four load-depth curves and indentations on quartz in sandstone.

Using the standard viscoelastic model and the extended viscoelastic model to fit the four nanoindentation strain-time curves of quartz in sandstone, fitting results are shown in Fig. 12 (a) and (b). And the fitting constants are given in Table 3.

![Standard and Extended Viscoelastic Models](image)

(a) The standard viscoelastic model  
(b) The extended viscoelastic model

**Fig. 12.** Fitting results of strain-time curves of quartz in sandstone by using the standard viscoelastic model and the extended viscoelastic model.

According to the fitting results in Fig. 12(a) and (b), both the standard viscoelastic model and the extended viscoelastic model can perfectly fit the nanoindentation data, with goodness above 0.99. However, like the obtained results in Section 4.2, fitting constants derived from the extended viscoelastic model tend to be mutually contradictory in Table 3, which are crossed out in this table.

**Table 3.** Mechanical parameters of quartz in sandstone.
As a result, the standard viscoelastic model was verified to be a suitable function to fit nanoindentation creep strain-time curves and calculate the creep parameters. The obtained hardness, elastic modulus and creep parameters are shown in Table 4. It also shows that the mechanical properties of quartz in sandstone are slightly larger than that of quartz in mudstone. For example, the mean elastic modulus of quartz in sandstone is 85.81 GPa and that in mudstone is 66.54 GPa. The reason for the smaller mechanical properties of quartz in mudstone is probably due to the influence of soft kaolinite as matrix in mudstone.

### Table 4. Mechanical parameters of quartz in sandstone.

| No. | $H_{IT}$/MPa | $E_{IT}$/ GPa | $E_i$/GPa | $\eta_i$/GPa/s | $\eta_v$/GPa/s |
|-----|--------------|--------------|-----------|----------------|----------------|
| 3-1 | 10416.00     | 86.55        | 1280.81   | 1884.09        | 37207.76       |
| 3-2 | 10440.00     | 87.99        | 1083.67   | 1364.14        | 42523.90       |
| 3-3 | 10078.00     | 85.38        | 1629.51   | 2010.00        | 35232.55       |
| 3-4 | 9912.00      | 83.32        | 1394.21   | 2331.46        | 30395.20       |

**Mean values:** 10211.50 85.81 1347.05 1897.42 36339.85

### 5 Conclusions and prospects

In this paper, the viscoelastic characteristics of kaolinite and quartz in mudstone were investigated by using the targeting nanoindentation technique. Conclusions are as follows.

1. The soft mudstone sample studied in this study are composed of kaolinite and quartz, in which kaolinite is soft and as matrix with hard quartz embedded in it.

2. For broken soft rocks that cannot provide intact standardized samples, the nanoindentation method can be used to investigate their mechanical properties by yielding stable mechanical parameters. Compared with the grid nanoindentation technique, the TNIT is efficient. This experimental technique can be realized by coupling nanoindentation technique with observing experimental methods, like XRD, SEM and EDS. Using this experimental method, the mechanical properties of constituents in geomaterials can be quickly obtained.

3. Nanoindentation creep strain-time curve reflects the viscoelastic properties of mudstone. The curve can be divided into two stages: transient stage and steady stage. The standard viscoelastic model consisting a viscous element and a Kelvin model in series can perfectly fit the nanoindentation creep strain-time curves of mudstone and provide reasonable rheological parameters. And the proposed model was also used to fit creep curves of quartz in sandstone with high goodness, which verify the nanoindentation creep model. Even though the extended viscoelastic can better fitting nanoindentation creep curves, it could yield unreasonable creep parameters.

4. The mechanical properties of quartz in mudstone and sandstone are a slightly different, which may be due to the influence of soft clay minerals in mudstone that soften the mechanical performance of quartz in it.

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