Results in Engineering 16 (2022) 100728

Contents lists available at ScienceDirect

Results in Engineering

journal homepage: www.sciencedirect.com/journal/results-in-engineering

Experimental study on the effect of thermal treatment on the mechanical properties of clay-rich rocks (mudstone)

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ARTICLE INFO

Keywords:
Thermal treatment
Clay-rich rocks
Mechanical properties
Tensile cracks
Shear cracks

ABSTRACT

Temperature is one of the most important environmental factors that affect rock engineering properties. The clay-rich rocks such as mudstone are abundant in the earth's crust and have a high affinity to water. So the behavior of these rocks differs from others after thermal treatment, which might force the water out of the clay. Therefore, we investigated the effect of thermal treatment on the mechanical properties of clay-rich rock, mudstone, under three temperatures; ambient temperature, 250, and 500 °C. Our results showed that thermal cracks begin to appear in the mudstone at a temperature between 500 and 600 °C. Several mechanical properties were measured in the lab; uniaxial compressive strength (UCS), elastic modulus (E), Poisson's ratio (ν), Brazilian tensile strength (BTS), fracture toughness of tensile (K_{IC}), and shear (K_{IIC}) cracks. Due to the drying of clay minerals through the dehydration and dihydroxylation processes, the following mechanical properties of mudstone significantly increased after treating the rock with a temperature of 500 °C; UCS, E, BTS, $K_{IC,}$ and K_{IIC} have improved by 91%, 39%, 16%, 41%, and 94%, respectively, while ν decreased by 80%. This reduction of ν, due to increasing the thermal treatment temperature, caused the behavior of mudstone to transform gradually from ductile to brittle material which means that the failure of such rocks, at this temperature, might be a sudden one. This research presents new insights into the effect of temperatures below 500 °C on clay-rich rocks for use in different engineering fields.

1. Introduction

There are many environmental factors to which rocks are exposed in nature, as these factors directly affect the strength and stability of the rock mass, geological structures, or any rock scattered in nature or used for building or industrial purposes. Thermal effects are among the most important environmental factors influencing the resistance of rocks. These effects can result from the change in temperature on the ground surface, the heat emitted from the earth's interior, the heat resulting from geothermal energy, underground coal gasification, nuclear engineering construction, etc. Clay-rich rocks such as mudstone are classified as sedimentary rocks that can form part of the oil and geothermal reservoirs [1–3]. Therefore, these rocks are affected by the geothermal temperature with depth and have different behavior from other rocks due to the clay's affinity to water. Another important application is the behavior of surface and near-surface rocks where during wildfires, these rock formations might be exposed to high temperatures that might cause landslides and rock falls [4–6]. An extensive study on the effect of temperature on the physical and mechanical properties is fundamental to better understanding the behavior of clay-rich rocks versus temperate changes.

The effect of thermal treatment on rock properties has been studied by many researchers. The studies and results differ according to the rock type, the temperatures to which the rock is exposed, and the rock properties. An overview of some studies related to the effect of thermal treatment on rock properties is shown in Table 1. This table includes different rock types, temperature ranges from ambient temperature to 1000 °C, and different rock properties. Most of the studies focused on the effect of thermal treatment on fracture toughness [7–13], especially fracture toughness under pure mode I. These studies generally indicated a decrease in fracture toughness under high-temperature of thermal treatment. Funatsu et al. [10] indicated that the K_{IC} of sandstone and tuff can be increased for a specific range of temperatures, this increase in K_{IC} was due to the drying of clays under high temperatures. Moreover, some studies have focused on the effect of temperature on elastic properties (E, ν), UCS, porosity, and wave velocity [13–16]. They concluded that

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https://doi.org/10.1016/j.rineng.2022.100728
Received 17 August 2022; Received in revised form 13 October 2022; Accepted 24 October 2022
Available online 30 October 2022

the rock behavior under compression is temperature-dependent. Due to the growth of thermal cracks, UCE, E, and wave velocity decrease with increasing temperature, while porosity and crack density increase. Ohnaka [17] and Odedra et al. [18] investigated the effect of temperature on the shear properties of granite and obtained the temperature dependence of the shear failure strength along the fault surface by a hyperbolic function of the temperature.

Clays are an important part of many rock formations in the Earth's crust, such as mudstone, limestone, sandstone, tuff, etc. Because of the unique properties of clay, it has many applications in the fields of agriculture, environment, and earth science. One of the most important applications is the natural geological engineering barrier for hazardous waste disposal including radioactive nuclear waste [19–21]. In general, the increase in the clay content leads to a decrease in the rock strength and can easily cause the deformation of rocks [22]. Some studies focused on the effect of temperature on clay-rich rocks such as claystone and shale. Tian et al. [23] investigated the physical and mechanical properties of claystone exposed to temperatures up to 1000 °C. They concluded that bulk density, deformation modulus, and compressive strength increase with increasing temperature to 800 °C. Guo et al. [24] studied the effect of bedding plane orientations and thermal treatment on shale and showed that the peak stress for bedding plane orientation of 0°, 45°' and 90° increases with temperature. Based on the inherent property of clay that increases its hardness when exposed to heat, and is widely used in the manufacture of ceramics, building materials, and archaeological artifacts [25,26]; the engineering properties of clay-rich rocks can be improved when they are thermally treated. Therefore, the focus of this research was to study the effect of thermal treatment on the basic engineering properties of mudstone that has a high content of clay minerals. The engineering properties of this study include Brazilian tensile strength, UCS, elastic modulus, Poisson's ratio, fracture toughness of tensile cracks under pure mode I, and fracture toughness of shear cracks under pure mode II. These tests and their results are important to understand the variation of mechanical properties of clay-rich rocks in response to thermal treatment.

2. Experimental study

The laboratory work in this study was conducted in the following sequence; the preparation of rock samples, thermal treatment in a furnace, preparation and analysis of microscopic sections, and testing. In this study, four different sample types were prepared to investigate the effect of thermal treatment on the mechanical properties of mudstone. The first type was Brazilian disks with dimensions of 50 mm in diameter and 25 mm in radius (H/D = 0.5) to measure Brazilian tensile strength (Fig. 1-a). The second type was cylindrical with 50 mm in diameter and 100 mm in height (H/D = 2) to measure uniaxial compressive strength, elastic modulus, and Poisson's ratio Fig. 1-b. The third type was cylindrical specimens that have two parallel notches of 50 mm in diameter and 100 mm in height to investigate the creation of shear cracks under pure shear stress. As shown in Fig. 1-c, these specimens have two notches with a length of D/2 = 25 mm and a ligament of 15 mm. The

Table 1
An overview of some studies related to the effect of thermal treatment on rock properties.

Rock type	Temperature range (T, °C)	Rock property	Observations	Reference
Limestone	20–600	UCS, E, ν, H, φ, V_p	• Decrease of UCS, E, H for $20 < T < 600$ °C. • Increase ν and Vp for $20 < T < 100$ °C, and then decrease them for $T > 100$ °C. • Increase of φ for $10 < T < 560$ °C, and then decrease of it for $560 < T < 600$ °C.	[13]
Sandstone	20–600	$K_{I/II,C}$, K_{IIC}	• $K_{I/II,C}$, K_{IIC} increased slowly for $20 < T < 100$ °C and decreased slowly for $100 < T < 500$ °C. • $K_{I/II,C}$, K_{IIC} decreased sharply for $500 < T < 600$ °C.	[7]
Gabbro Marble	20–600	K^d_{IC}	• Decrease of K_{IC} for $20 < T < 600$ °C.	[8]
Limestone	27–116	K_{IC}, $K_{I/II,C}$	• Increase of K_{IC} by about 25% with increasing T from 27 to 116 °C.	[9]
Sandstone Tuff	25–200	K_{IC}	• For sandstone, K_{IC} was almost constant for $25 < T < 125$ °C, and then started to increase for $T > 125$ °C. • For Tuff, K_{IC} decreased for $25 < T < 75$ °C, and then started to increase for $T > 75$ °C.	[10]
Basalt	AT - 750	K_{IC}	• Increasing T did not greatly affect the K_{IC} of the Etnean or Vesuvian basalt. • For Icelandic basalt, a marked increase of KIC was found at around T = 150 °C.	[11]
Siltstone	25–600	K_{IC}	• K_{IC} decreased slightly from ambient temperature 25–100 °C, then increased greatly from 100 to 125 °C, and then gradually declined from 125 to 600 °C.	[12]
Sandstone Dolerite	AT - 600	K_{IC}	• K_{IC} of sandstone and dolerite increased up to a temperature of 100 °C, and then decreased with a gradual increase in temperature.	[27]
Granite	100–600	K_{IC}	• Decrease in the load levels and fracture toughness of granite by increasing the temperature.	[28]
Granite	AT - 900	Crack density, φ, V_p, UCS	• Increase of crack density and φ with increasing T. • Decrease of V_p and UCS with increasing T.	[14]
Marble	AT - 1000	UCS, E	• For $T < 400$ °C, UCS and E fluctuated in different extents with the increase of T. • For $T > 400$ °C, UCS and E decreased linearly with the increase of T.	[15]
Calcarenite	105–600	φ, UCS, E, ν, Durability	• Increase of porosity • Decrease of UCS, Young's modulus, and durability • Increase of ν for T between 105 and 200 °C, slight decrease between 200 and 400 °C, high decrease after 400 °C.	[16]
Granite	AT - 500	UCS, V_P, fracture roughness, shear properties	• Increase of UCS by 37% at 200 °C and then decrease for T greater than 200 °C. • Linear decrease of *P*-wave velocity. • The degradation of fracture roughness is greater for high treatment temperatures. • The peak shear strength is not very affected For $T < 300$ °C, while it decreases by increasing the temperature above 300 °C.	[29]
Granite	−73 - 927	Shear properties	• The effect of T on shear strength is insignificant below 300 °C for dry granites (brittle regime). Above 300 °C, shear strength decreases due to the development of localized plastic deformation in the shear failure zone.	[17]
Granite	25–480	Shear properties	• Under 300 °C, wet and dry granite behaves as brittle material and shear strength is temperature insensitive. Above 400 °C, semi-brittle effects and ductility can be observed and shear strength decreases.	[18]

AT: Ambient temperature, UCS: Uniaxial compressive strength.
E: Elastic modulus, ν: Poisson's ratio, H: Hardness, φ: Porosity, V_P: *P*-wave velocity.
$K_{I/II,C}$: Fracture toughness under mixed-mode loading, K_{IC}: Fracture toughness under pure mode I, K_{IIC}: Fracture toughness under pure mode II.

fourth type was notched semicircular rock specimens with a diameter of 100 and a thickness of 40 mm. These specimens have a notch with a length of 50 mm to investigate the creation of tensile cracks under pure tension Fig. 1-d.

2.1. Thermal treatment

Rock samples were thermally treated at a constant temperature for 24 h in the furnace. The temperature increases gradually from the ambient to the desired temperature at an increasing rate of 4.2 °C per minute. Rock samples were exposed to temperatures from 100 to 700 °C while observing the appearance of thermal cracks on the specimen surface. At each temperature, the rock sample was placed in the furnace and then gradually exposed to the temperature until the desired temperature was reached. The sample remained for 24 h within the desired temperature. Then the furnace was turned off and the temperature gradually decreased until it returned to room temperature. The samples were gradually heated and cooled to avoid the thermal shock that could cause a sudden change in the rock properties. The original color of the samples at ambient temperature is beige. During heat treatment, rock samples acquire a gray color (cement color) and gradually become darker with increasing temperature. The notched semicircular specimens were thermally treated to monitor the growth of thermal cracks around the notch (see Fig. 2). Up to a temperature of 500 °C, no thermal cracks were observed on the specimen surface. Once the temperature reaches T = 600 °C, some thermal cracks developed between the upper and lower surfaces of the sample. At a temperature of T = 700 °C, an intersecting network of thermal cracks was observed on the sample surface. As a result of these experiments, the temperature at which thermal cracks begin to grow in mudstone was between 500 and 600 °C. Therefore, in this study the rock samples will be thermally treated at three main temperatures; the ambient temperature (T_1 = AT), the maximum temperature that the rock sample can sustain without growing thermal cracks (T_3 = 500 °C), a temperature between T_1 and T_3 which is set to be T_2 = 250 °C. After thermally treating the samples at each temperature, the mechanical properties of mudstone will be investigated.

2.2. Mineralogical composition

The microscopic thin section before and after thermal treatment was used to determine the composition of rock samples as shown in Fig. 3. White color in the initial sections indicates calcite, light to dark color indicates clay, and black color represents voids and pores. As shown in Fig. 3, calcite, clay, and void content were analyzed and displayed separately in red color. The proportions of the contents in the microscopic sections were determined depending on the analysis of grayscale, white, and black using the Image J program [30]. Where each content in the mudstone was converted to red color to make it easy to be distinguished from other contents, and then the percentage of content was determined by specifying the percentage of red color in each microscopic section. The microscopic section analysis shows that mudstone consists of 7% calcite, 92,75% clay, and 0.25% pores and voids. Based on these percentages (92.75% clay and 7% calcite), the geological classification of the used rock in this study is slightly calcareous mud/mudstone. Moreover, Fig. 3 shows the microscopic section at the three temperatures, and no color difference between the three microscopic thin sections under ambient, 250 °C, and 500 °C was observed. In addition, no thermal microscopic cracks were observed at 250 and 500 °C. As the growth of microscopic cracks under thermal treatment at a temperature greater than 500 °C increases the porosity of the rocks and significantly weakens their strength.

3. Experimental results

All mechanical tests were conducted by using the servo-hydraulic machine GOTECH AI-7000 L A (Fig. 6-a). The vertical displacement speed for this machine ranges between 0.001 and 250 mm/min and the maximum capacity is 20 tons. All tests in this study were conducted under the same displacement rate of 0.5 mm/min with a speed accuracy of ±0.5%.

3.1. Brazilian tensile strength (BTS)

Brazilian tensile strength of mudstone under three temperatures was determined depending on the standard test method for splitting tensile strength of intact rock core specimens ASTM D3967-16. The Brazilian disc was placed between the jaws of the loading device as shown in Fig. 4-a, as the loading increases the displacement was recorded. Three samples for each temperature were tested to validate and confirm the results. The displacement – load curves are shown in Fig. 4-b. This figure shows that at an ambient temperature of 25 °C the Brazilian disks fail at peak force between 5330 and 6984 N. After thermal treatment at a temperature of 250 °C, the failure force increases to 7432–9712 N. At a temperature of 500 °C, the failure force increases significantly in comparison to the ambient temperature to reach between 11,386 and 12,178 N. Brazilian tensile strength σ_t was then computed as follows:

$$\sigma_t = \frac{P_{max}}{\pi R T} \tag{1}$$

Where P_{max} is the failure force, R and T are the radius and thickness of the Brazilian disk, respectively. Fig. 4-c shows the Brazilian tensile strength as a function of the temperature of thermal treatment. As shown in the figure, the tensile strength of mudstone is temperature-dependent. By increasing the temperature from ambient temperature to 250 °C and 500 °C, the average value of tensile strength increases from 3.2 to 4.5 and 6,1 MPa, respectively. The results show that the thermal treatment at 250 and 500 °C improved the tensile strength by 41% and 91%, respectively.

3.2. Uniaxial compressive strength

The Uniaxial Compressive Strength (UCS) of the cylindrical rock specimens, after thermal treatment, was determined depending on the standard test method for UCS of intact rock core specimens [31]. Axial and lateral strain gauges were glued on the rock specimen to monitor vertical and horizontal strains and determine Young's modulus and Poisson's ratio (Fig. 5-a). These strain gauges are of type BQ 120-30 A A (120 Ohm in resistance and 30 mm in length). The stress-strain diagrams of the three rock specimens are shown in Fig. 5-b. The UCS (σ_c), elastic modulus (E), and Poisson's ratio (ν) were determined by using the stress-strain diagrams. Fig. 5-c shows that by increasing the temperature of thermal treatment from ambient temperature to 500 °C, UCS increased from 108.7 to 151.1 MPa and E from 42.7 to 49.4 GPa. As shown in the figure, the ν decreases from 0.377 to 0.303. This means that the UCS, E, have increased by 39%, and 16%, respectively, and ν decreased by 80%, due to increasing the temperature of thermal treatment from an ambient temperature to 500 °C. The increasing elastic modulus and decreasing Poisson's ratio means that the mudstone becomes more brittle with increasing the temperature of thermal treatment. Therefore, the transition from ductile to brittle behavior, for some types of rocks, can be achieved by thermally treating the rock. This affects the extent of elastic and plastic zones, strength, and all mechanical properties of the mudstone. In addition, Fig. 5-c shows that UCS, E, and ν have logarithmic relationships with the temperature of thermal treatment. With more extensive data, these relationships can be used to estimate the behavior of mudstone under unconfined compressive stress as a function of the temperature for engineering purposes, especially at great depths where the earth's crust is affected by any geothermal reservoir.

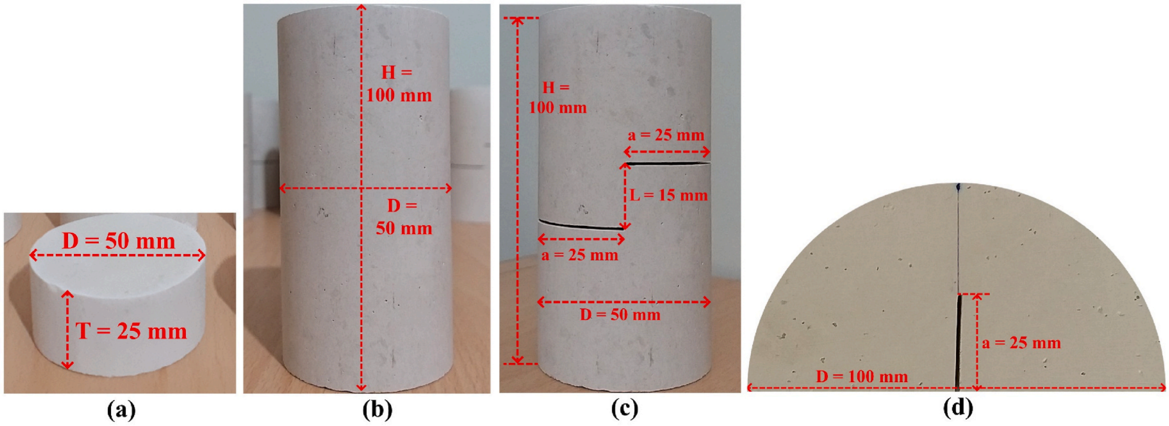

Fig. 1. Preparation of rock specimens, (a) Brazilian disc, (b) cylindrical specimen for UCS, (c) cylindrical specimen for shear cracks, (d) semicircular specimens for tensile cracks.

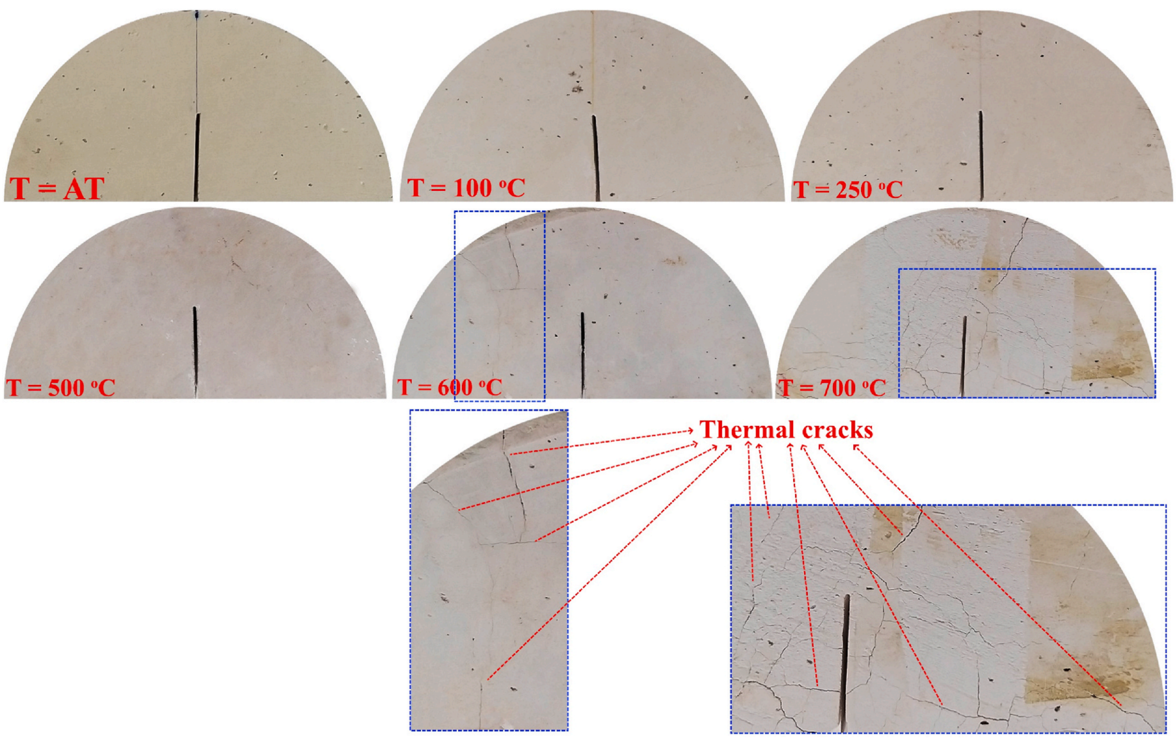

Fig. 2. Thermal treatment of rock samples.

3.3. *Three points bending test (tensile cracks)*

To investigate the thermal treatment effect on the fracture toughness of tensile cracks, the three points bending tests were conducted on semicircular specimens (SCB) according to ISRM's suggested method [32]. As shown in Fig. 6-a, the SCB specimen has a radius of 50 mm and is simply supported at a spacing (2 S) of 60 mm (S/R = 0.6). This specimen, as shown, has a notch with a length (a) of a = 25 mm (a/R = 0.5) and is loaded via the upper roller. According to the arrangement in Fig. 6-a, the SCB specimen is under pure tension and the fracture toughness of the tensile crack can be calculated by using:

$$K_{IC} = K^{I}_{Dim} \frac{P_{cr}}{2\,R\,T} \sqrt{\pi a} \tag{2}$$

Where K^{I}_{Dim} is the dimensionless stress intensity factor and can be calculated by a numerical procedure described by Alneasan et al. [33], Alneasan and Behnia [34], Alneasan, et al. [35]. They simulated numerically three points bending test on SCB specimen depending on the displacement discontinuity method and showed that for S/R = 0.6 and a/R = 0.5, $K^{I}_{Dim} = 4.18$. R, T, and (a) in equation (2) are the radius, thickness, and notch length of the SCB specimen. Meanwhile, the P_{cr} is the fracture load and is obtained from the peak of the displacement-load curve. The displacement-load curves for SCB specimens under ambient, 250 °C, and 500 °C temperatures are shown in Fig. 6-b. This figure shows that by increasing the temperature from ambient temperature to 250 °C and 500 °C, the average value of P_{cr} increases by 32% and 41%, respectively. Fracture toughness in equation (2) was calculated depending on the magnitudes of P_{cr} and the results are presented in Fig. 6-c. This figure shows a quadratic relationship between the fracture toughness and the temperature of thermal treatment. As expected and because the fracture toughness depends on the value of the fracture load as shown in Equation (2), increasing the treatment temperature from the ambient temperature to 250 °C and 500 °C, fracture toughens increase

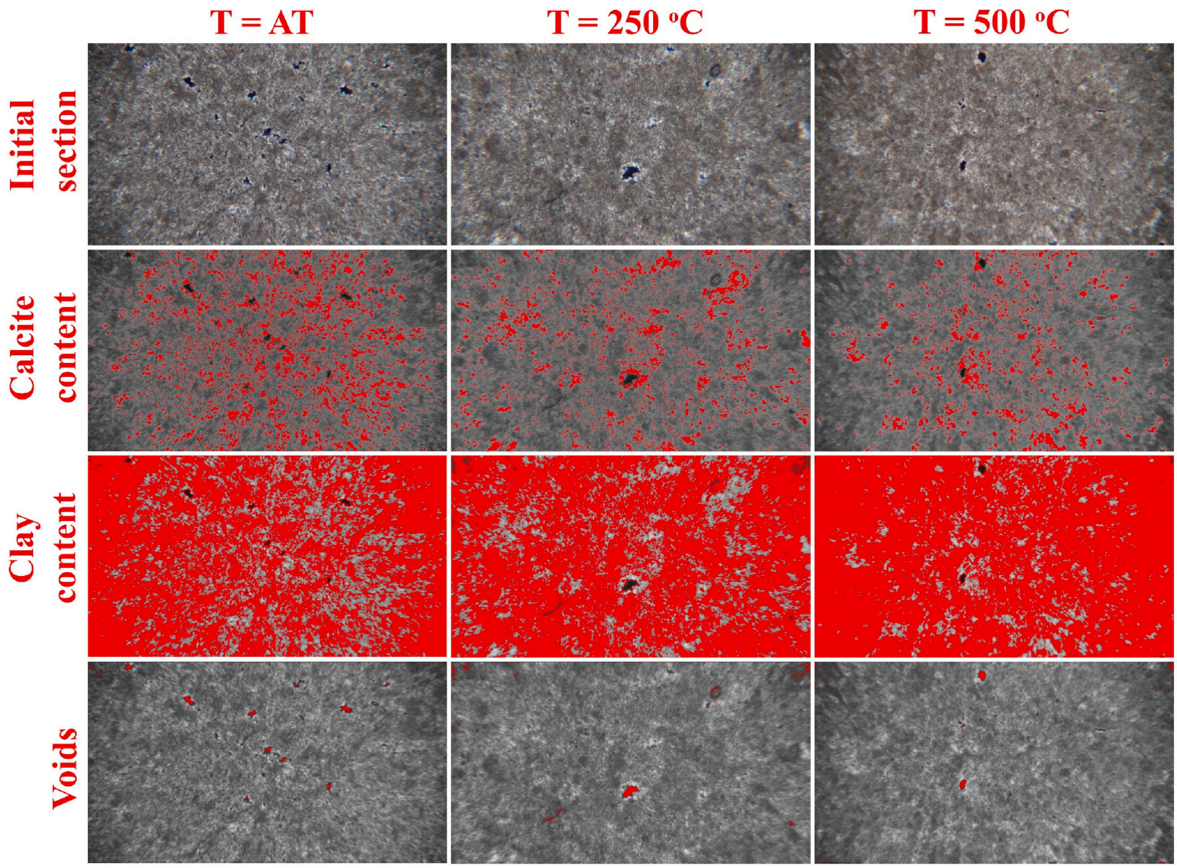

Fig. 3. Microscopic thin sections before and after thermal treatment.

by 32% and 41%, respectively.

3.4. *Short-notched core under compression (shear cracks)*

The main difference between this test and three points bending test is the mechanism of the formation of new cracks. As shown in Fig. 7-a, under three points bending test, a new crack grows under pure tensile stress as in hydraulic fracturing, blasting, joints, toppling, etc [36]. While in the short notched core under compression (SNCC), the new crack grows under pure shear stress (Fig. 7-a) as in the faulting process, earthquakes, rock slope sliding, etc. Fracture toughness (K_{IIC}) can be computed as follows [37]:

$$K_{IIC} = \left[2.3744\frac{L}{H} + 0.0192\right]\frac{P_{cr}}{2RL\sqrt{\pi a}} \tag{3}$$

Where L and H are the ligaments between two notches and the height of the specimen, respectively. P_{cr} is the fracture load and corresponds to the peak of displacement – the load curve. R and (a) are the radius and notch length, respectively. Displacement – load and fracture toughness – temperature diagrams are shown in Fig. 7-b and Fig. 7-c, respectively. These figures show that by increasing the temperature from ambient temperature to 250 °C and 500 °C, the average value of fracture load and fracture toughness increase by 33% and 94%, respectively. To compare between fracture toughness of tensile crack (K_{IC}) and fracture toughness of shear crack (K_{IIC}), the ratio between them was calculated and shown as a function of temperature in Fig. 7-c. This figure shows that under ambient temperature, K_{IIC} is 36.6 times greater than K_{IC}, while under 250 °C and 500 °C, K_{IIC} is 39.3 and 51.4 times greater than K_{IC}. This shows that the formation of cracks in the intact rock under shear loads is harder than that under tensile loads, and by thermally treating the mudstone, the formation of shear cracks becomes even more difficult (Fig. 7-c).

4. Discussion

The effect of thermal treatment on the mechanical properties of mudstone at temperatures between ambient temperature, 250 °C, and 500 °C was investigated in this study by using laboratory tests. Mechanical properties in this study included tensile and compressive strength, elastic modulus and Poisson's ratio, fracture toughness of tensile cracks under pure tension, and shear cracks under pure shear. In general, our results indicated a significant improvement in the mechanical properties of mudstone with increasing the temperature of thermal treatment. The behavior of rocks varies after thermal treatment based on their mineralogical composition and treatment temperature. Zhang et al. [13] showed that the mechanical properties (UCS, E, and ν) of limestone decreased by increasing treatment temperature from ambient temperature to 600 °C, while the porosity increased due to the escape of water and gases under high temperature, and due to the creation of new thermal cracks or expansion of preexisting cracks. Al-Shayea [38] indicated that for temperatures between ambient temperature and 116 °C, the K_{IC} of limestone increased by about 25%. Funatsu et al. [10] investigated the effect of thermal treatment on the fracture toughness of sandstone and tuff and indicated that the fracture toughness of sandstone was constant for temperatures less than 125 °C, and it increased for temperatures between 125 °C and 200 °C. While for the tuff, the K_{IC} decreased for temperature between 0 °C and 75 °C and increased for temperature between 75 °C and 200 °C. They illustrated that the decreasing of K_{IC} in tuff was due to the effect of microcracks, but the increase of K_{IC} in sandstone and tuff was due to the drying of the clay minerals. Mahanta et al. [27] investigated the effect of thermal treatment on three rock types (two types of sandstone and one type of dolerite) at temperatures between room temperature and 600 °C. They

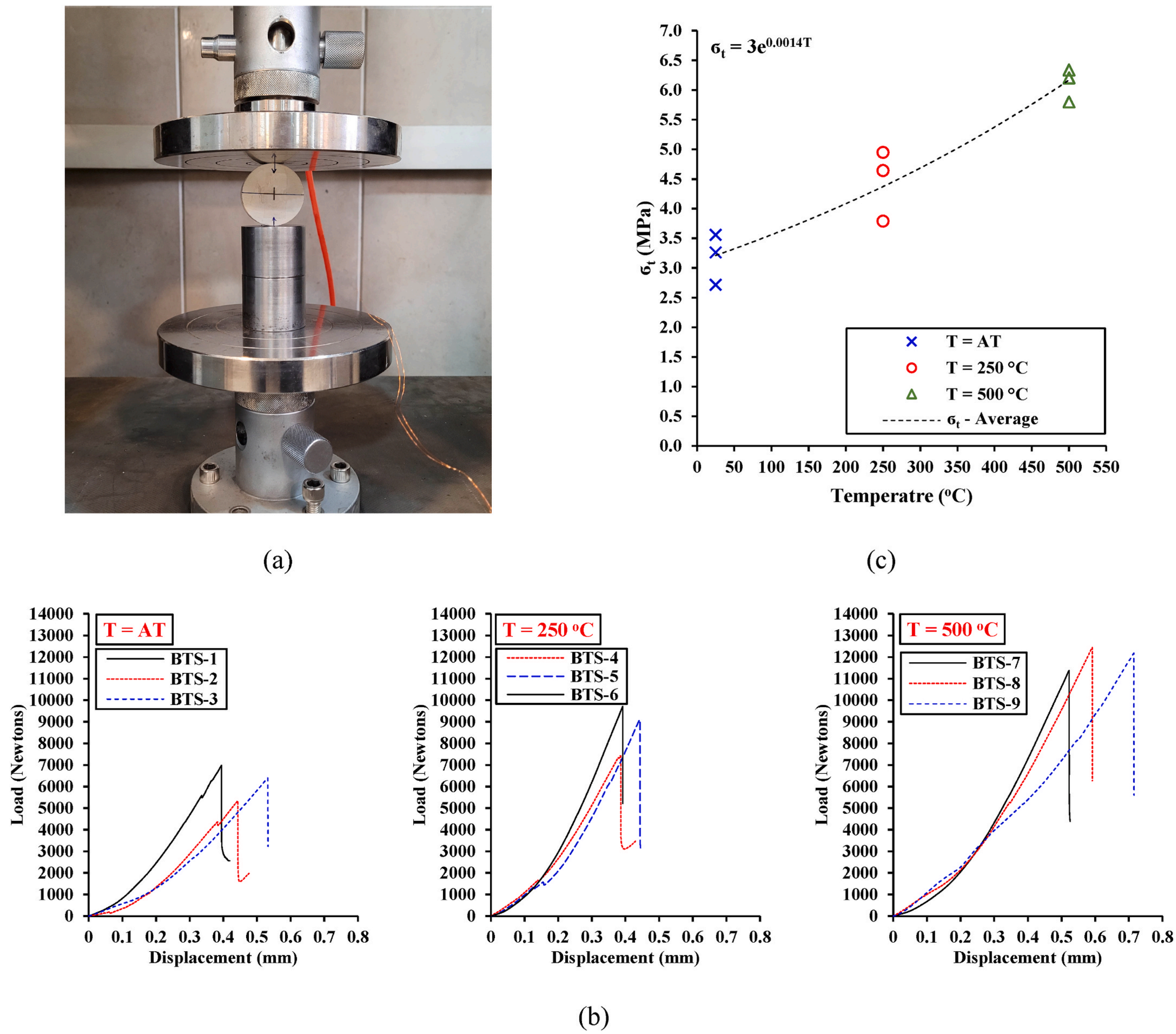

Fig. 4. Brazilian splitting test, (a) Brazilian disc between the jaws of the loading device, (b) displacement – load curves, (c) Brazilian tensile strength versus temperature of thermal treatment.

concluded that for treatment temperatures less than 100 °C, K_{IC} increases with increasing the temperature, while for treatment temperatures between 100 °C and 200 °C, K_{IC} decreases.

According to the mineralogical composition of mudstone used in this study, the main constituent minerals of this rock type are clays (92.75%) and calcite (7%). Due to the high percentage of clays, it can be said that the used mudstone in this study is a clay containing calcium compounds such as calcium carbonate ($CaCO_3$), dolomite ($CaMg(CO_3)_2$), calcium bicarbonate ($Ca(HCO_3)_2$), and magnesium carbonate ($Mg(CO)_3$). Thermal treatment mainly contains two stages: the first stage includes the decomposition of original minerals, while the second stage includes the formation of new silicates. At temperatures between 200 °C and 300 °C, calcium bicarbonate decomposes to calcium carbonate ($CaCO_3$), H2O, and CO_2 (equation (3) [13]). Dolomite ($CaMg(CO_3)_2$) decomposition to calcium oxide (CaO), magnesium oxide (MgO) and CO_2 can occur at a temperature between 300 and 500 °C (about 350 °C) according to equation (4) [39]. At temperatures between 500 and 600 °C, magnesium carbonate decomposes to Magnesium oxide (MgO) and CO_2 (equation (5) [13]). At temperatures 700–900 °C, calcium carbonate can be decomposed into calcium oxide (CaO) releasing carbon dioxide (equation (6) [40]).

$$Ca(HCO_3)_2 \rightarrow CaCo_3 + H2O + CO_2 \ (200 \text{ to } 300\ ^{\circ}C) \tag{3a}$$

$$CaMg(CO_3)_2 \leftrightarrow CaO + MgO + 2CO_2 \ (300 \text{ to } 500\ ^{\circ}C) \tag{4}$$

$$MgCO_3 \rightarrow MgO + CO_2 \ (500 \text{ to } 600\ ^{\circ}C) \tag{5}$$

$$CaCO_3 \rightarrow CaO + CO_2 \ (700 \text{ to} + 900\ ^{\circ}C) \tag{6}$$

Clays mainly consist of clay minerals, subordinate quartz, feldspars, anatase, carbonates, and goethite [26]. During thermal treatment, clay elements undergo the following processes: dehydration, dehydroxylation, decomposition and formation of new phases, and vitrification. Trindade et al. [26] represented a mineralogical transformation in calcite-rich clay induced by thermal treatment at temperatures between 300 °C and 1100 °C and concluded that quartz, calcite, illite, kaolinite,

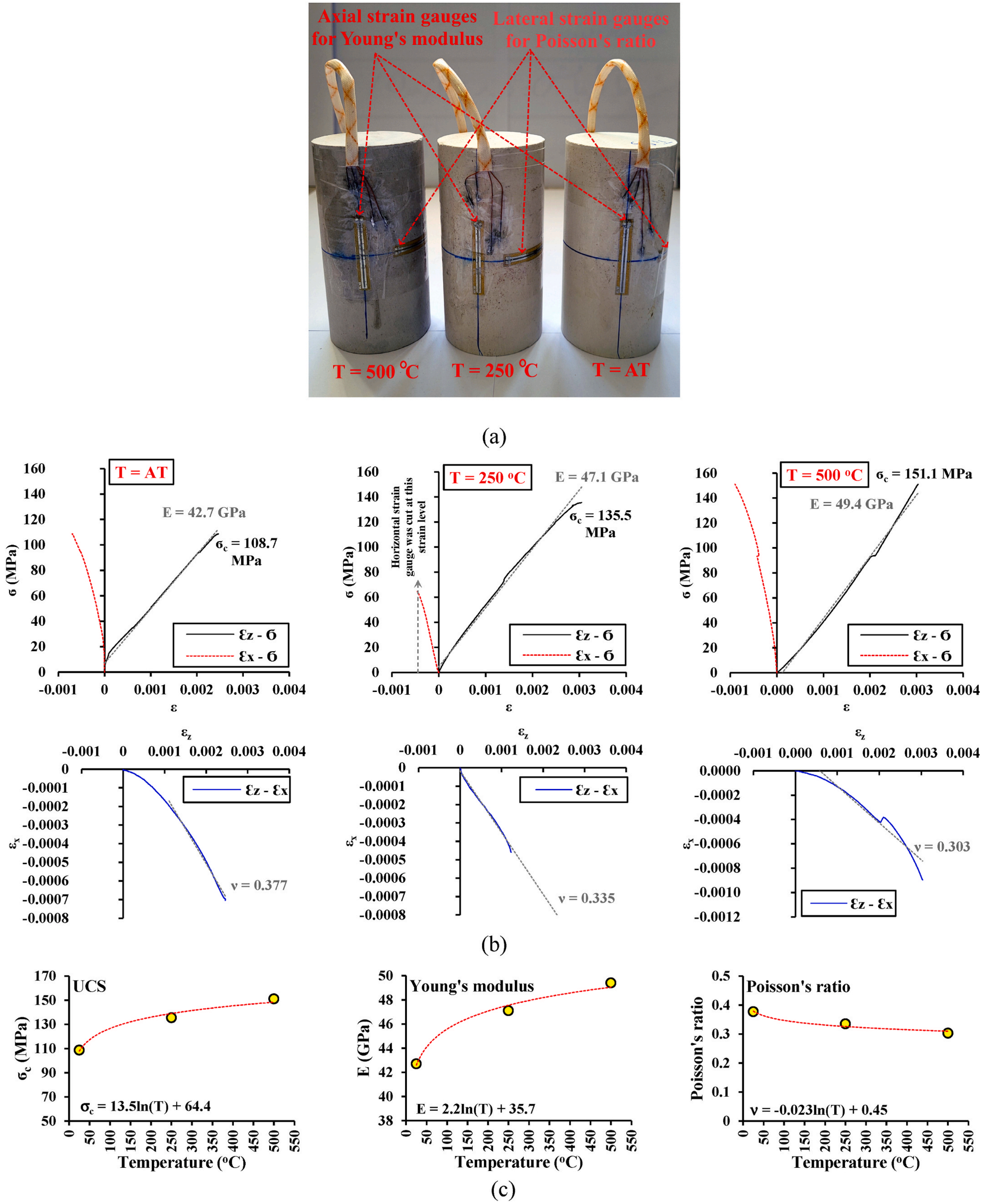

Fig. 5. Uniaxial compressive strength, (a) cylindrical specimens of mudstone after thermal treatment with axial and lateral strain gauges, (b) strain–stress curves, (c) UCS, E, and ν as a function of the temperature of thermal treatment. Where ε_z-σ corresponds to the vertical strain-stress curve, ε_x-σ corresponds to the lateral strain-stress curves, ε_z-ε_x corresponds to the vertical-lateral strain curve.

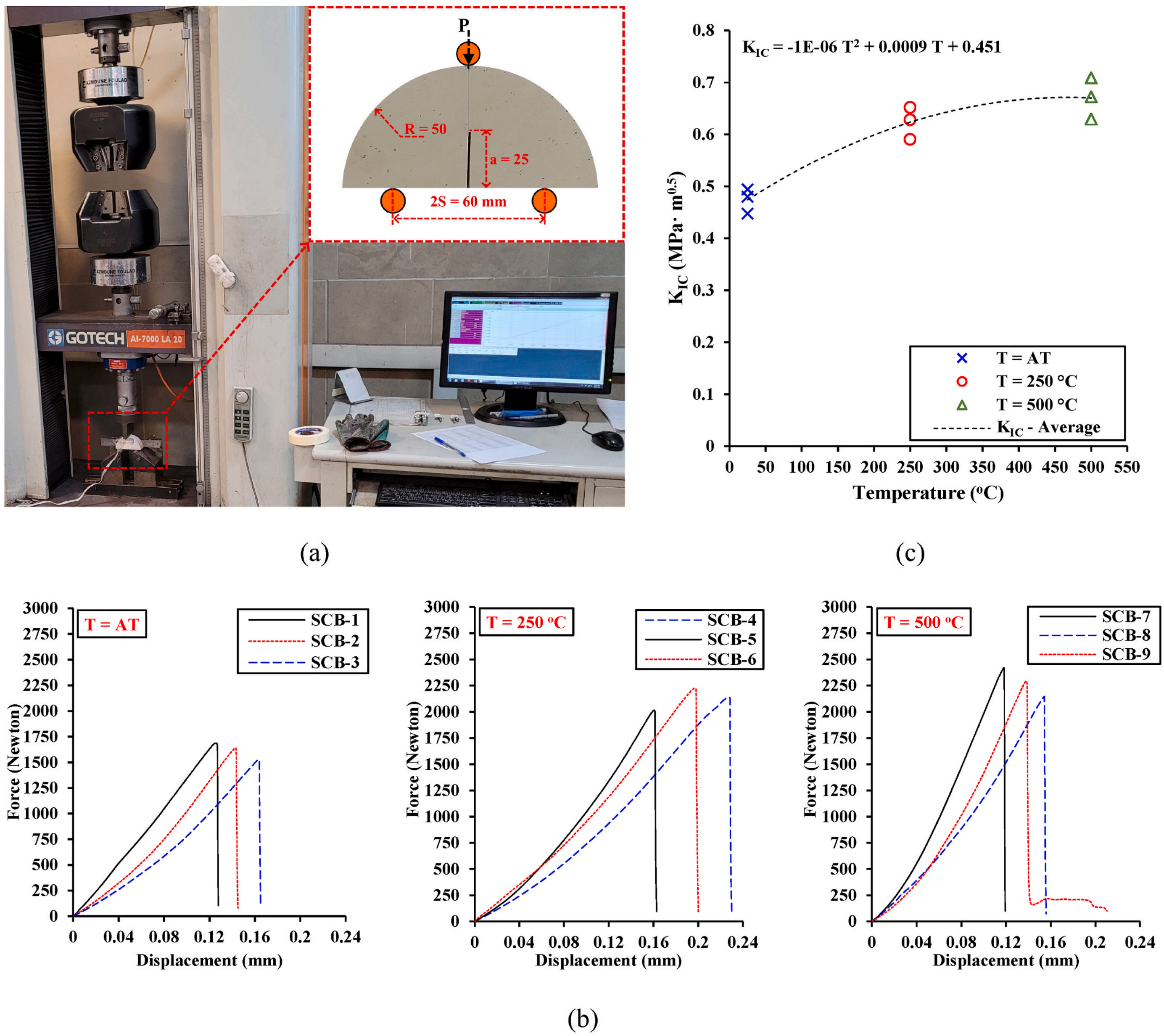

Fig. 6. SCB specimens under three points bending test, (a) loading device, (b) displacement – load curves, (c) fracture toughness versus temperature of thermal treatment.

anatase, plagioclase disappeared at temperatures after 1100, 1000, 800, 400, 700, 600 °C, respectively. K-feldspar and goethite disappeared at temperatures less than 300 °C. Some other components appeared after specific temperatures, hematite formed at temperatures greater than 400 °C, gehlenite, wollastonite, larnite, γ-alumine, and (Ca, K) sulfate appeared at temperatures larger than 800 °C. D'Elia et al. [41] concluded that heating increases the silica-to-alumni ratio and reduces the content of calcite, silica, and alumni. Houhou et al. [42] investigated the effect of thermal loading on the microstructure of the compacted clays and discussed some hypotheses for the effect of temperature on clays. By increasing the temperature, the thickness of the diffuse double layer decreases, and as a result, the particles get closer together and the contact between them increases, which in turn leads to an increase in the density and an improvement in the properties of clay [43]. In addition, under the effect of temperature, the particles are rearranged due to the expansion of the diffuse double layer, this also leads to an increase in the contact points between the particles and thus improves the properties of the clay [44].

Based on the above, the mechanical properties of clay-rich rocks such as mudstone significantly improve by increasing the temperature of thermal treatment. Our results indicated that by increasing the temperature from ambient temperature to 500 °C, tensile strength, UCS, elastic modulus, Poisson's ratio, fracture toughness of tensile crack (K_{IC}) and shear crack (K_{IIC}) have improved by 91%, 39%, 16%, 80%, 41%, and 94%, respectively. The increasing mechanical properties of mudstone by increasing the temperature are mainly due to the removal of water, decomposition of the preexisting clays, and formation of new clay minerals. The results of this study agree with experimental results in clay-rich rocks such as claystone under high temperatures up to 1000 °C. Where Tian et al. [23] indicated an increase in the mechanical properties of claystone in a temperature range between ambient temperature and 800 °C. These and our findings were supported by some researchers such as Funatsu et al. [10], Mahanta, et al. [27], Funatsu, et al. [45] who indicated an increase in the fracture toughness with increasing the temperature of thermal treatment to a certain limit. They concluded that this increase in the fracture toughness results from the hardening of clay

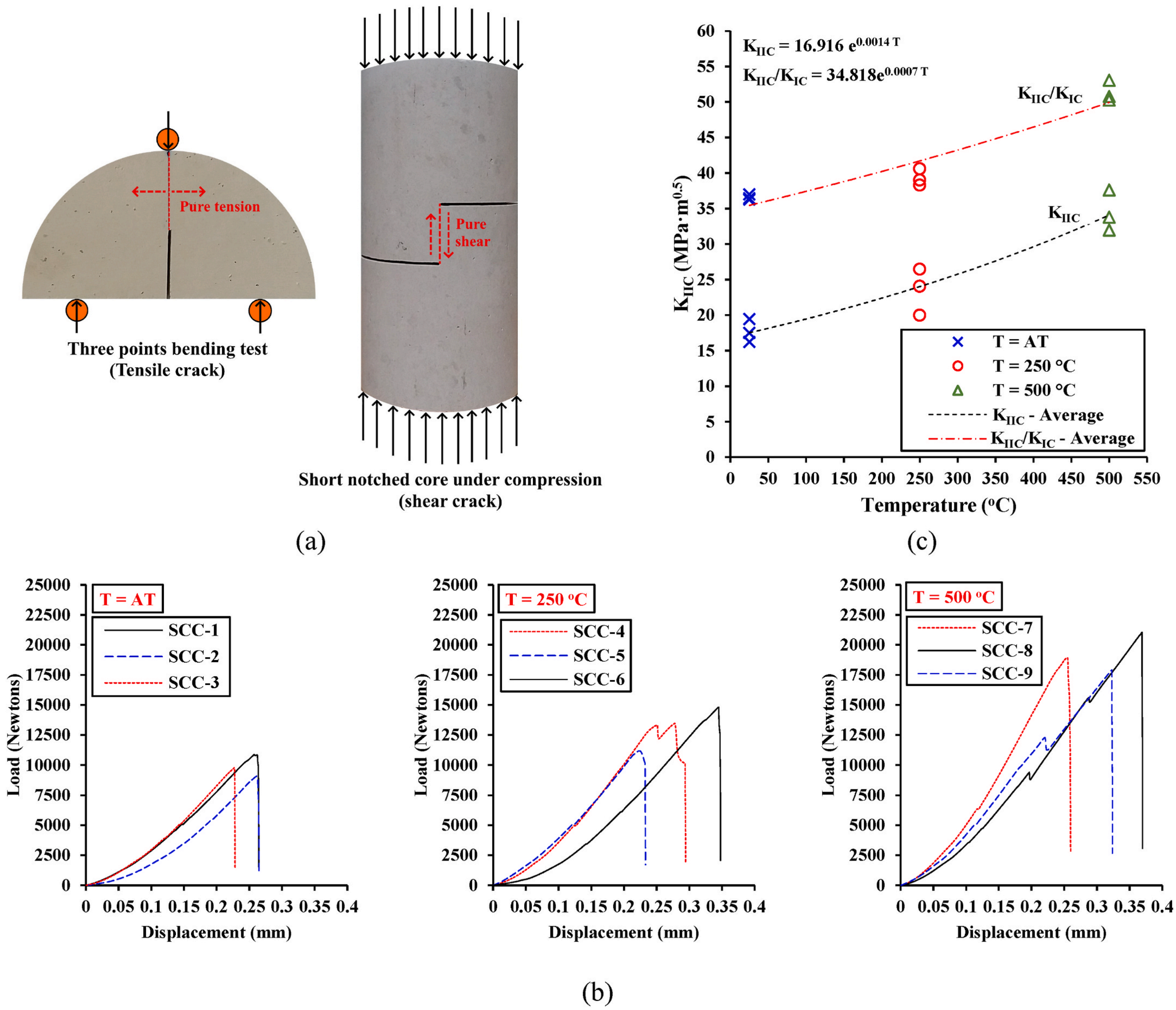

Fig. 7. Short notched core under compression (SNCC), (a) the mechanism of formation of the new crack in SCB vs SNCC, (b) displacement – load curves, (c) fracture toughness as a function of temperature. Where K_{IIC}/K_{IC} is the fracture toughness ratio which corresponds to the ratio of pure mode II to pure mode I fracture toughness.

minerals which is due to the dehydration or dry-caking of the interlayers and the absorptive water above 100 °C, which increases the coefficient of friction between mineral particles, leading to an increase in the fracture toughness, above 100 °C. Water is removed through two stages, dehydration and dihydroxylation. The dehydration process includes the ejection of adsorbed, pore, and interlayer water. This process takes place at temperatures between 100 and 200 °C. The dihydroxylation process takes place at temperatures between 500 and 1000 °C. In this process, hydroxyl ions leave the crystal structure of the clay minerals promoting new cracks to grow, propagate and coalesce as observed in our experiments, where some thermal cracks appeared in the samples between 500 and 700. At temperatures between 200 and 500 °C, phase transition, water evaporation and some chemical and physical reactions can occur due to the decomposition and expansion of some minerals, especially calcium compounds. Changes in the mineralogical composition of the mudstone during thermal treatment are summarized in Fig. 8. These changes were summarized depending on the previous discussion. As shown in this figure, a temperature of 500 is approximately a turning point in the behavior of the mudstone used in this study. Before this point, the mechanical properties of the mudstone are improved due to the drying of clay minerals. While after this point, a decrease in mechanical properties is expected due to the growth of some thermal cracks.

According to the effect of thermal treatment on the rock behavior under unconfined compressive stress, by increasing the temperature and drying of clay minerals through dehydration and dehydroxylation processes, Young's modulus increases and Poisson's ratio decreases, and the behavior of mudstone can transform gradually from ductile to brittle behavior due to water leaving the mudstone at high temperature. Under ambient temperature, the presence of pore and interlayer water in mudstone increases its ductility. Gradual removal of water through the dehydration process causes an increase in the brittleness degree and rock resistance, until the temperature at which thermal cracks begin to appear (500–600 °C in this study). The transition from brittle to ductile behavior occurred with increasing temperature, but it should be remembered that the deformation-controlled compression test was conducted on samples cooled at room temperature. Drying of clay minerals occurs during the first two stages of thermal treatment, the gradual heating stage, and the constant temperature stage. But this process is not reversible, which means that clay minerals do not recover the lost water between their layers during the gradual cooling process, especially since there are minerals that appear and others disappear during thermal treatment as shown in Fig. 8. Therefore, the transition from ductile to brittle behavior and the gradual decrease of Poisson's ratio is not reversible during the gradual cooling stage.

It should be mentioned that only the total percentage of clays (as 92.75%) and the total percentage of calcite (as 7%) are determined in this study. For more accuracy in future works, the type of clay minerals, interlayer minerals in clays and calcium compounds should be determined by using other scientific methods such as X-ray diffraction method (XRD). Therefore, the effect of temperature on calcium compounds and clay minerals was analyzed and discussed in this study based on results of the similar studies. Where we showed in Fig. 8 how the changes occurred in the calcium compounds and clay minerals during thermal treatment. In addition, at a temperature greater than 500 °C, which will result in a lot of thermal cracks on mudstone, the failure mechanical behavior of rock sample will happen obvious changes compared with before 500°. Therefore, in future works, it is recommended to study the effect of thermal treatment on the mechanical properties of mudstone at temperatures greater than 500 °C.

5. Conclusions

The effect of thermal treatment on the mechanical properties of clay-rich rocks such as mudstone was investigated in this study. Four types of rock specimens were prepared; cylindrical specimens to measure UCS, elastic modulus, and Poisson's ratio, notched cylindrical specimens to investigate the creation of shear cracks, Brazilian disks for tensile strength, and notched semi-circular specimens to investigate the effect of temperature on tensile cracks. Firstly, some specimens were thermally treated at temperatures from 100 to 700 °C and the results showed that the temperature at which thermal cracks begin to grow varies between 500 and 600 °C. Therefore, the basic tests were conducted under ambient temperature, 250 °C, and 500 °C.

The results indicated one behavior for the effect of temperature on the mechanical properties of the mudstone. By increasing temperature, all mechanical proeprteis have improved due to drying of clay minerals through dehydration and dehydroxylation processes. By increasing temperature from ambient temperature to 250 and 500 °C, tensile strength increased by 41% and 91%, respectively. Unconfined

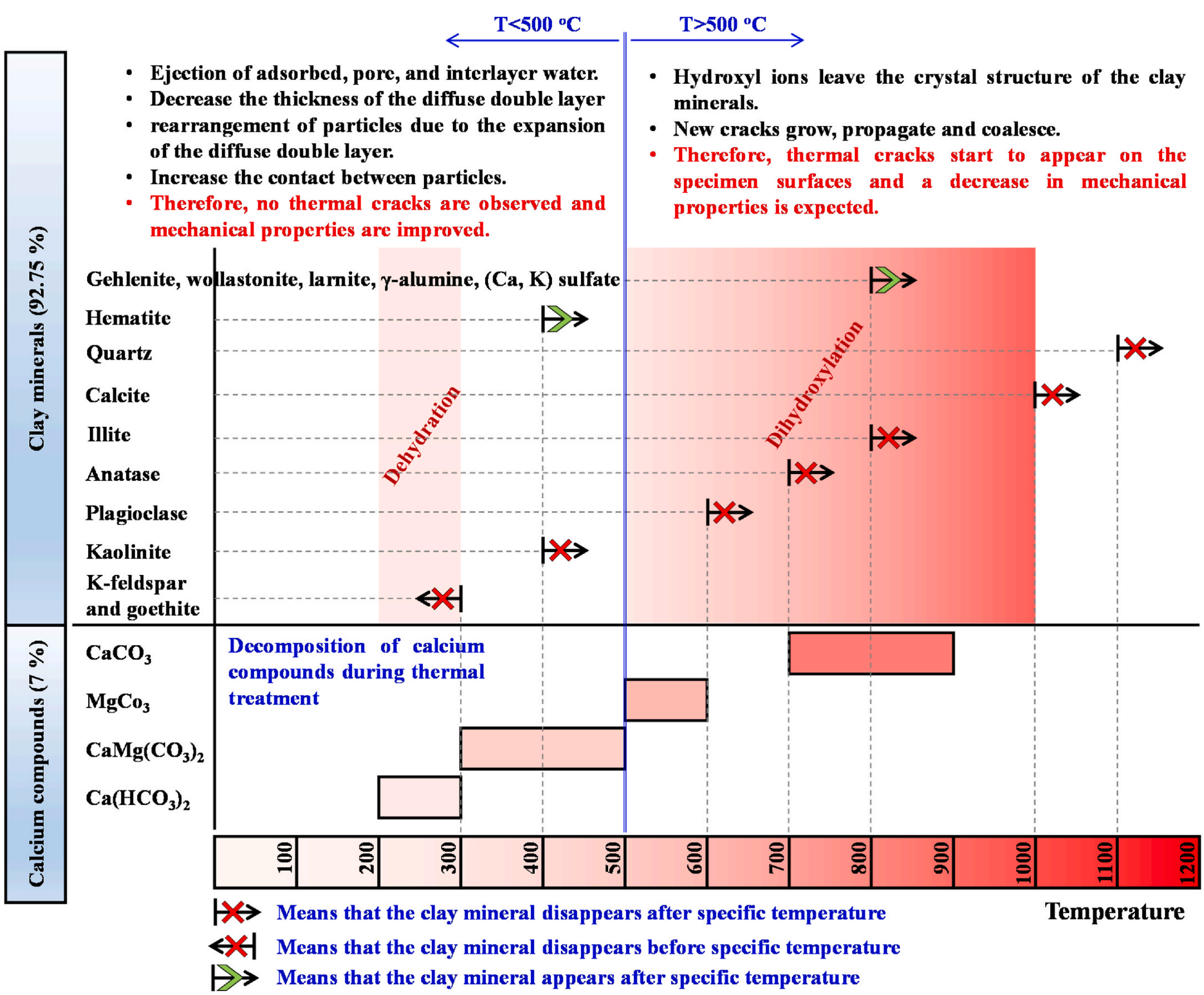

Fig. 8. Changes in the mineralogical composition of the mudstone during thermal treatment.

compressive stress tests showed that the rock behavior under uniaxial stress (UCS, E, and ν) is significantly affected by thermal treatment, by increasing the temperature of thermal treatment from an ambient temperature to 500 °C, UCS and E increase from 108,7–151.1 MPa and 42.7–49.4 GPa, respectively, while the Poisson's ratio decreases from 0.377 to 0.303. Which indicate a gradual transition in rock behavior from ductile to brittle one. Taking into account temperature-fracture behavior in rock mass for fractures under tensile and shear loads, results indicated that fracture toughness of tensile K_{IC} and shear K_{IIC} cracks are directly proportional to the temperature of thermal treatment. But the increasing rate of fracture toughness of shear cracks was greater than that of tensile cracks. Therefore, the formation of cracks in rock masses under shear loads is more difficult than that under tensile loads and by increasing the temperature of thermal treatment of mudstone, the formation of shear cracks becomes more difficult.

The results of this study are of particular interest to be used in the rock community to better understand the effect of temperature on the mechanical properties of clay-rich rocks such as mudstone. Especially in deep rock engineering practices and deep underground excavations. Where it is important to analyze stresses induced by thermal effects to enhance stability and safety of underground structures.

6. Authorship statement

Conceptualization: Mahmoud Alneasan, Abdel Kareem Alzo'ubi.; Data curation: Mahmoud Alneasan.; Formal analysis: Mahmoud Alneasan, Abdel Kareem Alzo'ubi.; Funding acquisition: Abdel Kareem Alzo'ubi.; Investigation: Mahmoud Alneasan, Abdel Kareem Alzo'ubi.; Methodology: Mahmoud Alneasan, Abdel Kareem Alzo'ubi.; Project administration: Abdel Kareem Alzo'ubi.; Resources: Abdel Kareem Alzo'ubi, Mahmoud Alneasan.; Software: Mahmoud Alneasan, Abdel Kareem Alzo'ubi.; Supervision: Abdel Kareem Alzo'ubi.; Validation: Abdel Kareem Alzo'ubi, Mahmoud Alneasan.; Visualization: Mahmoud Alneasan, Abdel Kareem Alzo'ubi.; Writing - original draft: Mahmoud Alneasan.; Writing - review and editing: Abdel Kareem Alzo'ubi.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

No data was used for the research described in the article.

Acknowledgment

We would like to thank Abu Dhabi University and the ORSP for funding this project (19300529, 19300528). We also would like to extend our thanks to Azmoneh Foulad Consulting Engineers[1] for permission to use facilities in this company's rock mechanics lab under the supervision of Eng. Mohsen Aghababaei[2] and Eng. Behzad Nateghpour.[3]

References

[1] J. Cui, X. Yuan, S. Wu, R. Zhang, S. Jin, Y. Li, Rock types and reservoir characteristics of shahejie formation marl in shulu sag, jizhong depression, bohai bay basin, J. Earth Sci. 32 (2021) 986–997.

[2] X. Fu, J. Wu, J. Shou, X. Wang, J. Zhou, T. Zhang, Y. Guo, Tight oil reservoir characteristics of lacustrine mixed marlstone in the palaeogene shahejie formation of shulu sag, bohai bay basin, Oil Gas Geol. 40 (2019) 78–91.

[3] G. Zou, H. Zeng, F. Gong, C. Yin, S. Li, S. Peng, Z. Xu, Effects of temperature and water saturation on the elastic moduli of sandstone and mudstone from the Lufeng geothermal field: experimental results and theoretical model, Geophys. J. Int. 230 (2022) 1147–1165.

[4] S.H. Cannon, J.E. Gartner, M.G. Rupert, J.A. Michael, A.H. Rea, C. Parrett, Predicting the probability and volume of postwildfire debris flows in the intermountain western United States, Bulletin 122 (2010) 127–144.

[5] D.M. Staley, A.C. Tillery, J.W. Kean, L.A. McGuire, H.E. Pauling, F.K. Rengers, J. B. Smith, Estimating post-fire debris-flow hazards prior to wildfire using a statistical analysis of historical distributions of fire severity from remote sensing data, Int. J. Wildland Fire 27 (2018) 595–608.

[6] J.V. de Graff, A.J. Gallegos, The challenge of improving identification of rockfall hazard after wildfires, Environ. Eng. Geosci. 18 (2012) 389–397.

[7] G. Feng, Y. Kang, F. Chen, Y.-w. Liu, X.-c. Wang, The influence of temperatures on mixed-mode (I+ II) and mode-II fracture toughness of sandstone, Eng. Fract. Mech. 189 (2018) 51–63.

[8] Z. Zhang, J. Yu, S. Kou, P.-A. Lindqvist, Effects of high temperatures on dynamic rock fracture, Int. J. Rock Mech. Min. Sci. 38 (2001) 211–225.

[9] N. Al-Shayea, K. Khan, S. Abduljauwad, Effects of confining pressure and temperature on mixed-mode (I–II) fracture toughness of a limestone rock, Int. J. Rock Mech. Min. Sci. 37 (2000) 629–643.

[10] T. Funatsu, M. Seto, H. Shimada, K. Matsui, M. Kuruppu, Combined effects of increasing temperature and confining pressure on the fracture toughness of clay bearing rocks, Int. J. Rock Mech. Min. Sci. 41 (2004) 927–938.

[11] M. Balme, V. Rocchi, C. Jones, P. Sammonds, P. Meredith, S. Boon, Fracture toughness measurements on igneous rocks using a high-pressure, high-temperature rock fracture mechanics cell, J. Volcanol. Geoth. Res. 132 (2004) 159–172.

[12] J.-p. Zuo, H.-p. Xie, F. Dai, Y. Ju, Three-point bending test investigation of the fracture behavior of siltstone after thermal treatment, Int. J. Rock Mech. Min. Sci. 70 (2014) 133–143.

[13] W. Zhang, Q. Sun, S. Zhu, B. Wang, Experimental study on mechanical and porous characteristics of limestone affected by high temperature, Appl. Therm. Eng. 110 (2017) 356–362.

[14] L. Griffiths, M. Heap, P. Baud, J. Schmittbuhl, Quantification of microcrack characteristics and implications for stiffness and strength of granite, Int. J. Rock Mech. Min. Sci. 100 (2017) 138–150.

[15] S. Liu, J. Xu, Study on dynamic characteristics of marble under impact loading and high temperature, Int. J. Rock Mech. Min. Sci. 62 (2013) 51–58.

[16] V. Brotons, R. Tomás, S. Ivorra, J. Alarcón, Temperature influence on the physical and mechanical properties of a porous rock: san Julian's calcarenite, Eng. Geol. 167 (2013) 117–127.

[17] M. Ohnaka, A shear failure strength law of rock in the brittle-plastic transition regime, Geophys. Res. Lett. 22 (1995) 25–28.

[18] A. Odedra, M. Ohnaka, H. Mochizuki, P. Sammonds, Temperature and pore pressure effects on the shear strength of granite in the Brittle-Plastic Transition Regime, Geophys. Res. Lett. 28 (2001) 3011–3014.

[19] L. Liu, H. He, M. Dyck, J. Lv, Modeling thermal conductivity of clays: a review and evaluation of 28 predictive models, Eng. Geol. 288 (2021), 106107.

[20] A. Gens, L.d.N. Guimaraes, A. Garcia-Molina, E. Alonso, Factors controlling rock–clay buffer interaction in a radioactive waste repository, Eng. Geol. 64 (2002) 297–308.

[21] M. Villar, J. García-Siñeriz, I. Bárcena, A. Lloret, State of the bentonite barrier after five years operation of an in situ test simulating a high level radioactive waste repository, Eng. Geol. 80 (2005) 175–198.

[22] L. He, Q. Meng, D. Li, Y. Zhang, L. Ren, Influence of dietary fiber sources on the physiological and behavioral stress responses of Greylag geese (Anser anser), J. Appl. Poultry Res. 25 (2016) 389–395.

[23] H. Tian, M. Ziegler, T. Kempka, Physical and mechanical behavior of claystone exposed to temperatures up to 1000 C, Int. J. Rock Mech. Min. Sci. 70 (2014) 144–153.

[24] Y. Guo, L. Huang, X. Li, J. Chen, J. Sun, Experimental investigation on the effects of thermal treatment on the physical and mechanical properties of shale, J. Nat. Gas Sci. Eng. 82 (2020), 103496.

[25] M. Maggetti, Phase analysis and its significance for technology and origin, in: Archaeological Ceramics, 1982, pp. 121–133.

[26] M.J. Trindade, M. Dias, J. Coroado, F. Rocha, Mineralogical transformations of calcareous rich clays with firing: a comparative study between calcite and dolomite rich clays from Algarve, Portugal, Appl. Clay Sci. 42 (2009) 345–355.

[27] B. Mahanta, T. Singh, P. Ranjith, Influence of thermal treatment on mode I fracture toughness of certain Indian rocks, Eng. Geol. 210 (2016) 103–114.

[28] T.Y. Guo, L.N.Y. Wong, Z. Wu, Microcracking behavior transition in thermally treated granite under mode I loading, Eng. Geol. 282 (2021), 105992.

[29] F. Meng, J. Song, L.N.Y. Wong, Z. Wang, C. Zhang, Characterization of roughness and shear behavior of thermally treated granite fractures, Eng. Geol. 293 (2021), 106287.

[30] M.D. Abràmoff, P.J. Magalhães, S.J. Ram, Image processing with ImageJ, Biophot. Int. 11 (2004) 36–42.

[31] D. ASTM, Standard Test Method for Unconfined Compressive Strength of Cohesive Soil, ASTM standard D, 2017, p. 2938.

[32] M.D. Kuruppu, Y. Obara, M.R. Ayatollahi, K. Chong, T. Funatsu, ISRM-suggested method for determining the mode I static fracture toughness using semi-circular bend specimen, Rock Mech. Rock Eng. 47 (2014) 267–274.

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[33] M. Alneasan, M. Behnia, R. Bagherpour, Analytical and numerical investigations of dynamic crack propagation in brittle rocks under mixed mode loading, Construct. Build. Mater. 222 (2019) 544–555.

[34] M. Alneasan, M. Behnia, Strain rate effects on the crack propagation speed under different loading modes (I, II and I/II): experimental investigations, Eng. Fract. Mech. 258 (2021), 108118.

[35] M. Alneasan, A.K. Alzo'ubi, M. Behnia, O. Mughieda, Experimental observations on the effect of thermal treatment on the crack speed and mode I and II fracture toughness in brittle and ductile rocks, Theor. Appl. Fract. Mech. (2022), 103525.

[36] A.K. Alzo'ubi, Effect of strength properties on displacement mag-magnitude and fracturing amount inside a large open pit prone to toppling, Journal of GeoEngineering 14 (2019) 65–76.

[37] Y. Xu, W. Yao, G. Zhao, K. Xia, Evaluation of the short core in compression (SCC) method for measuring mode II fracture toughness of rocks, Eng. Fract. Mech. 224 (2020), 106747.

[38] N. Al-Shayea, Comparing reservoir and outcrop specimens for mixed mode I–II fracture toughness of a limestone rock formation at various conditions, Rock Mech. Rock Eng. 35 (2002) 271–297.

[39] G. Cultrone, F.J.C. Rosua, Growth of metastable phases during brick firing: mineralogical and microtextural changes induced by the composition of the raw material and the presence of additives, Appl. Clay Sci. 185 (2020), 105419.

[40] G. Cultrone, C. Rodriguez-Navarro, E. Sebastian, O. Cazalla, M.J. De La Torre, Carbonate and silicate phase reactions during ceramic firing, Eur. J. Mineral 13 (2001) 621–634.

[41] A. D'Elia, D. Pinto, G. Eramo, L. Giannossa, G. Ventruti, R. Laviano, Effects of processing on the mineralogy and solubility of carbonate-rich clays for alkaline activation purpose: mechanical, thermal activation in red/ox atmosphere and their combination, Appl. Clay Sci. 152 (2018) 9–21.

[42] R. Houhou, M. Sutman, S. Sadek, L. Laloui, Microstructure observations in compacted clays subjected to thermal loading, Eng. Geol. 287 (2021), 105928.

[43] I. Towhata, P. Kuntiwattanaku, I. Seko, K. Ohishi, Volume change of clays induced by heating as observed in consolidation tests, Soils Found. 33 (1993) 170–183.

[44] R. Plum, M. Esrig, Some temperature effects on soil compressibility and pore water pressure, Highway research board special report 103 (1969) 231–242.

[45] T. Funatsu, M. Kuruppu, K. Matsui, Effects of temperature and confining pressure on mixed-mode (I–II) and mode II fracture toughness of Kimachi sandstone, Int. J. Rock Mech. Min. Sci. 67 (2014) 1–8.