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**MECHANICAL BEHAVIOUR OF AN AUSTRALIAN MUDSTONE SUBJECTED TO HEATING  
TREATMENT AT HIGH-TEMPERATURES**

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## **MECHANICAL BEHAVIOUR OF AN AUSTRALIAN MUDSTONE SUBJECTED TO HEATING TREATMENT AT HIGH-TEMPERATURES**

### **ABSTRACT**

This study falls in the context of underground coal fires (UCF) where burning coal can elevate the temperature of a rock mass in excess of thousand degrees. The objective of the research was to experimentally investigate the change in mechanical behavior of a rock when subjected to temperatures up to 900 °C. Several blocks of an Australian mudstone were collected from a mining site in New South Wales, Australia. X-ray diffraction and thermal-gravimetric analysis were conducted on powdered rock in order to determine the critical temperatures at which the main mineralogical changes occur. Then, cylindrical specimens were prepared from intact rock blocks for unconfined compression tests. These rock specimens were then treated by heating in a temperature-controlled furnace at temperatures varying from 100 °C to 900 °C. The heating process consisted of two stages: raising temperature with very slow rate of 1 °C per hour followed by a holding period of 24 hours and then a slow cooling process to room temperature. The compression tests were carried out on treated rock specimens to characterize the change in compressive strength. The macroscopical study was complemented by microstructural investigations via optical microscope, scanning electron microscope (SEM) and mercury intrusion porosimetry (MIP). Results show that the unconfined compressive strength of mudstone decreases with increasing heating temperature up to 450 °C, beyond which it increases. This can be explained by the combined effects of micro cracks and mineral thermal reactions caused by heating process.

### **KEYWORDS**

Mudstone, Heating Treatment, High Temperature, Mercury intrusion porosimetry, SEM

### **INTRODUCTION**

Underground coal fires (UCFs) are a serious environmental problem of global dimension. These fires, which can result from human activity (in most cases through mining activities) or occur naturally, are problematic for many reasons including loss of valuable non-renewable resources, mine safety, damage to infrastructure and damage to the natural environment (Stracher & Taylor, 2004; Sinha & Singh, 2005; Kuenzer et al., 2007). This is why underground coal fires have been of great concern to scientists and engineers (Voigt & Rüter, 2005). UCFs are characterized by very slow burning kinetics due to the limited availability of oxygen underground and very elevated temperature at the core of the fire because of surrounding ground preventing heat dissipation. Temperatures at combustion center of UCFs of up to 1300 °C have been reported in the literature (Stracher & Taylor, 2004). The continuous combustion of the coal seam typically leads to a progressive collapse of the overburden soil/rock mass, that is left unsupported, and to the formation of large cracks, which provide new air circulation paths allowing the fire to propagate (Ide et al., 2010).

It has been recognized that the existing sets of discontinuities tend to influence the roof collapse and patterns of crack propagation. However, the effect of heat generated by the fire on the mechanical properties of the rock mass has been overlooked. The present experimental work was initiated to investigate the influence of high temperatures on the mechanical behaviour of some Australian rocks, that could possibly be associated with underground coal fires (Liu et al. 2012a, b).

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Many studies have been carried out over the last several decades on the impact of high temperatures on the physical and mechanical properties of rocks and this was in different engineering fields such as underground coal gasification or nuclear waste disposal (Dmitriev, 1972; Somerton 1992; Sygala et al. 2013, to name few). It has been found that apart from the external stress, two key factors govern the changes in rocks when exposed to high temperatures: mineral thermal expansion and mineral thermal reaction (Tian et al., 2012). The former may induce great internal thermal stresses as a consequence of the differences in thermal-expansion characteristics of minerals, resulting in thermal cracking and strength degradation (David et al. 1999; Keshavarz et al., 2010). On the other hand, mineral thermal reaction may result in some new bonding agents, which can strengthen the rocks (Luo and Wang, 2011). The predominance of one phenomenon over the other depends on the mineral composition of rocks and thermal loading conditions (temperature and its change rate, heating and cooling cycle) as well as external stress state, the mechanical response of the heated rocks would be different as summarized by Zhang et al. 2009; Ranjith et al., 2012; Brotóns et al. 2013. Most studies focused on the change at macroscopic level with only a few studies attempting to investigate the fundamental physic-chemical mechanisms from a microscopic perspective (Hajpál, 2002; Keshavarz et al. 2010; Yavuz et al. 2010). This study investigates the change in unconfined compressive strength of a sedimentary mudstone subjected to heating treatment up to 900 °C. Microstructural analysis techniques (MIP and SEM) were combined to thermal-gravimetric analysis (TGA) to enhance the understanding of the mechanisms of heating treatment on the mechanical response of the mudstone tested. In order to try to simulate the very slow heating process of rocks in UCFs, a relatively slow heating process was used: the heating period proceeded at a rate of 1 °C per hour and was followed by a holding period of 24 hours, before cooling was initiated. This was also deemed to reduce the differential thermal stresses inside the rock generated by rapid heating and thereby the development of microcracks.

### Material

A sedimentary mudstone was collected from a mining site in Hunter Valley, New South Wales, Australia. It is referred to as HV mudstone. The basic physical properties of the intact (i.e. no heat treated) HV mudstone and its main mineralogical composition are presented in Table 1. The presence of dominant minerals such as Quartz and Muscovite was confirmed by microscopic imaging analysis on a thin section of HV mudstone (Fig. 1). The microscopic images also indicate that the maximum particle size is less than 100 µm and there are some evident bedding planes existing in the mudstone (Fig. 1b).

Table 1. Some physical properties and dominant minerals of HV mudstone

Specific gravity ( $G_s$ )	Bulk density ( $g/cm^3$ )	Initial void ratio ( $e$ )	Mineralogical composition from Semi-quantitative XRD analysis
$2.6567 \pm 0.002$	$2.38 \pm 0.03$	$0.11 \pm 0.01$	Quartz (66%), Muscovite (12%), Albite (7%), Kaolinite (9%), Anorthite (4%), Vermiculite (< 1%)

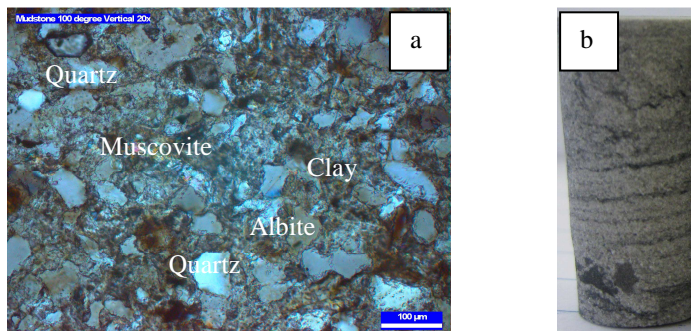


Figure 1- (a) Microscopic view of minerals in a thin section and (b) Macroscopic view of HV mudstone

## EXPERIMENTAL METHODS AND TESTING PROGRAM

### Experimental Methods

#### Thermal-Gravimetric Analysis

To highlight the mineralogy changes of HV mudstone in response to heating, thermal-gravimetric analysis was performed on the oven-dried mudstone powder with particle size below 75  $\mu\text{m}$ . The tests were run under Argon environment whilst raising the temperature of 20  $^{\circ}\text{C}$  to 1100  $^{\circ}\text{C}$  at a rate of 5  $^{\circ}\text{C}/\text{min}$ . The mass loss and heat flow were recorded during the heating process, which enables the determination of the critical temperatures at which the dominant mineral thermal reactions take place.

#### Specimen preparation and mechanical testing

The rock samples were cored perpendicular to the bedding planes (as shown in Fig.1b) with a diameter of 12 mm and a height of 24 mm. Note that these dimensions comply with the ASTM standard D-4543 (2008), which states that the diameter should be at least ten times the size of the biggest particles. With the particles smaller than 0.1 mm, 12 mm diameter satisfies this criterion. Such small dimensions helped limiting the variability of the material. The specimens were first dried in an oven at a temperature of 100  $^{\circ}\text{C}$  (reference state). Then, the dried specimens were subjected to heating treatment at different target temperatures (300, 450, 600, 750 and 900  $^{\circ}\text{C}$ ) by using a temperature-controlled furnace. The heating treatment consisted of a slow heating period from 20  $^{\circ}\text{C}$  to target temperatures at a rate of 1  $^{\circ}\text{C}/\text{hour}$  and a holding period of 24 hours followed by a cooling process at a rate of 25  $^{\circ}\text{C}/\text{hour}$  back to 20  $^{\circ}\text{C}$ . Note that the rock specimens were periodically weighted during the heating process, in order to monitor the mass loss. Uniaxial compression tests were conducted on the heated specimens according to ASTM standard D7012-2014. The loading rate of these tests was 0.8 mm/hour.

#### Mercury intrusion porosimetry (MIP) and Scanning electron microscope (SEM) observations

The MIP analyses were conducted using a Micromeritics AutoPore (IV 9500) with the mercury pressure incrementally increased to the maximum value of the apparatus (228 MPa), which corresponds to an equivalent entrance pore diameter of approximately 0.0065  $\mu\text{m}$ . As for the SEM observations using a Philips XL30 SEM, Oxford ISIS EDS, the specimens were coated by gold powder in order to obtain the better contrast of SEM image prior to SEM observations. Both MIP tests and SEM observations were carried out on the specimens after heating treatment

#### Testing Program

Table 2 summarizes all the tests performed in this study, including TGA, UCS tests and microstructural analysis.

Table 2 Testing program (TGA, UCS test, MIP and SEM)

Specimen heating temperature ( $^{\circ}\text{C}$ )	TGA	Unconfined compression test	MIP	SEM observation
100	1	2	1	1
300	-	1	-	-
450	-	1	1	1
600	-	2	1	1
750	-	2	-	-
900	-	1	1	1

## RESULTS AND DISCUSSION

### Variation of Mass Loss Ratio with Temperature

Fig. 2 presents the mass loss ratios (in percent of initial mass) of HV mudstone in response to an increase in temperature, which were respectively determined by thermal-gravimetric analysis (20 to 1100 °C) and from heating treatment using a temperature-controlled furnace (100 to 1200 °C). In order to more accurately determine the critical temperatures at which the dominant mass loss occurred, the differential thermal-gravimetric (DTG) evolution was also shown in Fig.2, which was obtained by simply differentiating the mass loss ratio curve.

It can be noticed that the mass loss ratio from TGA slightly decreases over the temperature range below 400 °C with the sharpest drop located between 400 and 600 degrees.

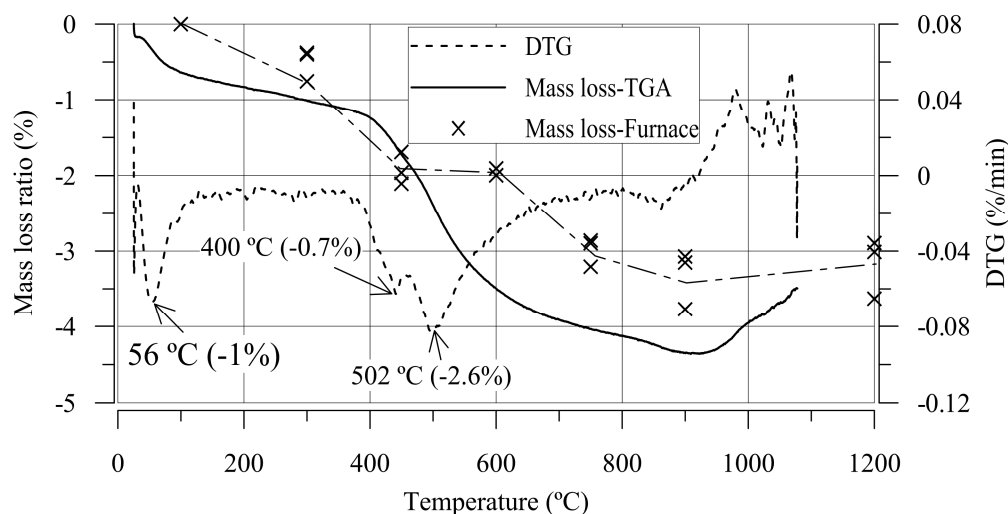


Figure 2. Comparison of mass loss ratio of HV mudstone determined from TGA and heating treatment using temperature-controlled furnace

Table 3. Thermal reactions of the dominant minerals of HV mudstone (Dmitriev 1972; Somerton 1992)

Dominant minerals	Critical temperatures (°C)	Thermal reactions
Quartz (66 %)	573	$\alpha$ - $\beta$ transition
	1470	Fusion
Muscovite (12%)	125	Loss of hygroscopic water
	450-650	Loss of structural water
	850-900	Lattice destruction
Kaolinite (9%)	455-642	Loss of structural water
	960	Lattice restructuring
Albite (7%)	1500-1550	Fusion
Anorthite (4%)	1100-1250	Fusion

Interestingly, a small proportion of mass gain can be noticed when the heating temperature exceeds 900 °C. A similar trend was observed for the specimens burnt in the furnace although there exists a clear offset of about 1%. This offset is believed to be due to the type of specimen: the TGA requires powdered rock with a higher surface area of material exposed to the heat, resulting in higher mass loss ratio. To understand the mechanism related to phenomenological mass loss, possible temperature alterations on the dominant minerals of the mudstone tested is summarized in Table 3 as shown by Dmitriev 1972; Somerton 1992. We can clearly see that the main mass loss is likely to be associated with the loss of

structural water from the muscovite and kaolinite, which are characterized by the DTG peak of 502 °C (Fig. 2). These reactions can possibly produce some bonding agents enabling to strength the mudstone as shown by Hajpál & Török (1998) and Hajpál (2002).

### Variation of Unconfined Compressive Strength (UCS) with Temperature

Figure 3 shows the evolution of UCS and corresponding axial strain at peak strength of the burnt HV mudstone with increase in temperature. Interestingly, the evolution is non-monotonic: the UCS first decreases from 100 °C to 450 °C before increasing over the reference strength (i.e. at 100 degrees). The values of strain at failure are more scattered and no clear trend can be defined. The mechanical response of the burnt mudstones may be explained by the change in their mineralogical composition and their microstructure. The former is governed by the thermal reactions of two dominant minerals (muscovite and kaolinite), which may create some cementation effects during heating at temperature above 450 °C, as discussed above. For the temperatures below 450 °C, the significant reduction in UCS may be imputable to the creation of micro cracks by heating treatment as suggested in some studies (e.g. Wolf 2006). This will be further verified by the microstructural analysis discussed in the following sections.

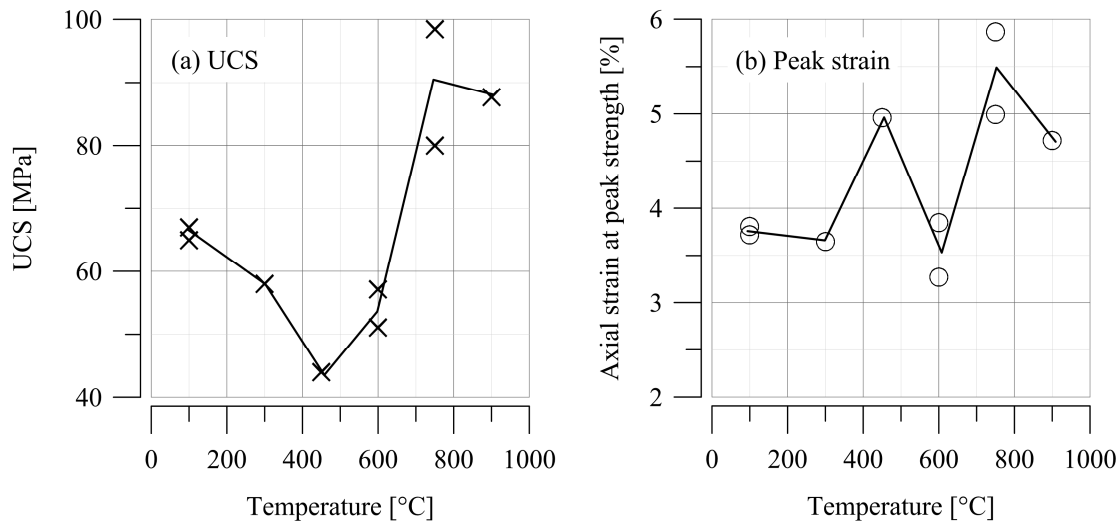
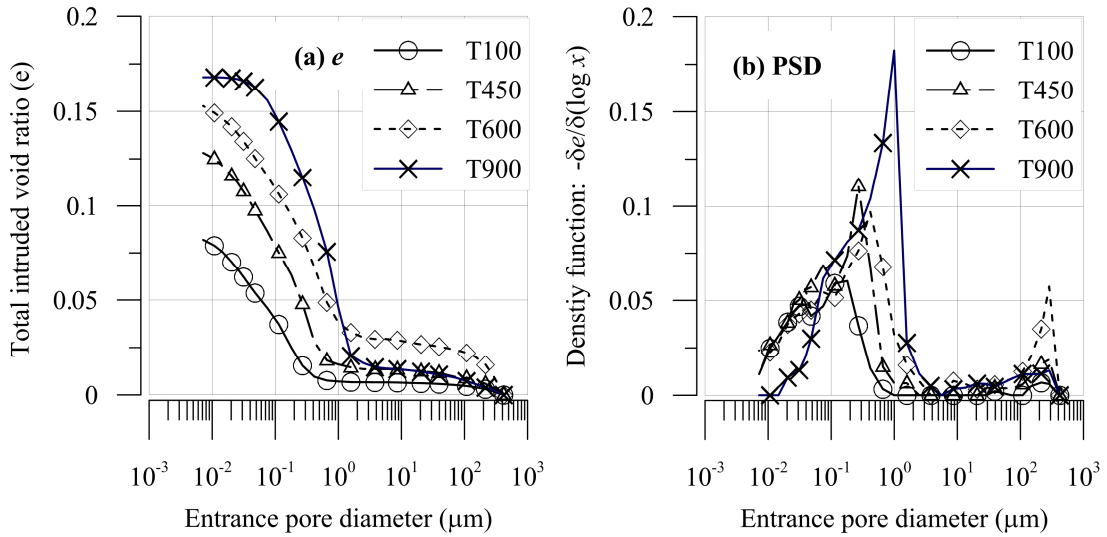


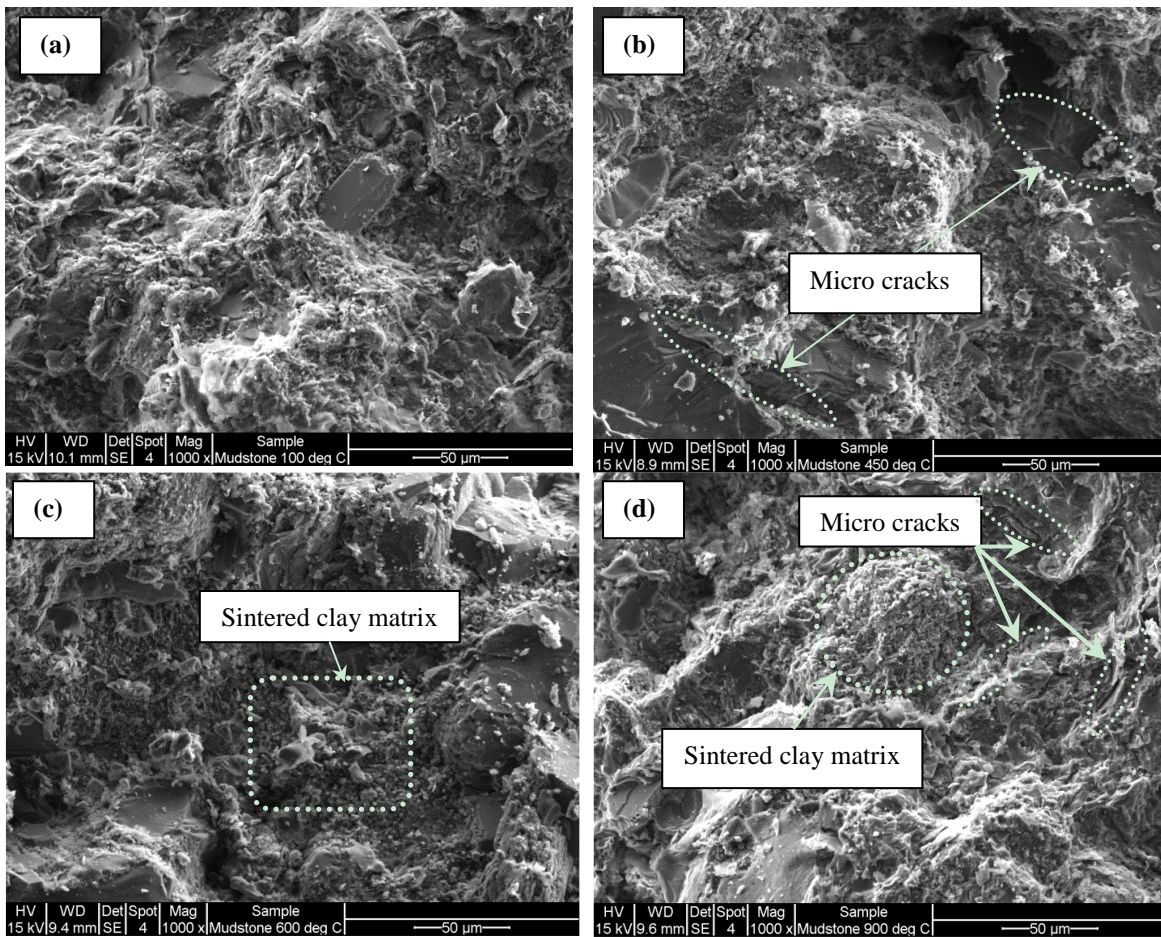
Figure 3. Variation of UCS and axial strain at peak strength of burnt HV mudstone with temperature

### Change in Microstructure with Temperature

Figure 4 presents the MIP results obtained on the mudstone specimens after heating treatment at temperatures of 100, 450, 600 and 900 °C while the corresponding SEM images appear in Figure 5. Figure 4 clearly shows that the total void ratio intruded by mercury intrusion increases by a factor 2 (from 0.08 to 0.17) as the heating temperature rose from 100 to 900 °C. This corresponds to a shift in dominant pore size from 0.1 μm to 1 μm. These changes can be explained by two basic mechanisms: (1) the differential thermal expansion of the different minerals (Table 3), (2) thermal reactions of the dominant minerals (Muscovite and kaolinite), resulting in the loss of structural water. The first one may induce the creation of microcracks inside the mudstone that are visible on the SEM images (Fig. 5b, 5d), leading to the strength degradation. As for the thermal reactions of the two minerals, it tends to create the sintered phases shown in Fig. 5c and 5d, which results in a strengthening effect. The predominance of one mechanism over the other governs the final strength changes of the mudstones in response to heating treatment. For instance, the second mechanism seems to play a more important role in strength changes at temperatures over 450 °C, at which the onset of dominant thermal reactions took place as shown in Table 3 and Fig. 2. This explains the non-monotonic UCS evolution and the transition from the decrease to the increase in UCS.



**Figure 4.** MIP results of four mudstone specimens burnt at temperatures of 100, 450, 600 and 900 °C: (a) Variation of total mercury intrusion void ratio with entrance pore diameter (b) Pore size distribution



**Figure 5.** SEM images obtained on four mudstone specimens burnt at temperatures of 100°C (a), 450 °C (b), 600 °C (c) and 900 °C (d)



## CONCLUSIONS

This study aimed at investigating the thermal effects on UCS of a sedimentary mudstone collected from a Hunter Valley site close to an existing underground coal seam fire. The compression tests were combined to thermal-gravimetric analysis and microstructural analysis that were employed to understand the mineralogical and microstructural changes associated with increasing temperatures. The tests showed that the UCS changes non-monotonically with temperature and this can be explained by two dominant mechanisms: differential thermal expansion of the minerals (Quartz, Muscovite, kaolinite and Albite) and thermal reactions of muscovite and kaolinite over the temperature range of interest. The predominance of one mechanism over the other governs the final strength changes of the mudstones in response to heating treatment. The first mechanism tends to induce microcracking, and increase in porosity as evidenced respectively by SEM observations and mercury intrusion porosimetry (Fig. 5, Fig. 4).

The second mechanism consists of dehydration and dehydroxylation processes, in other words, the loss of strongly adsorbed water and the structural water. The dehydroxylation occurs at temperatures above 450 °C and can create some sintered phases, which results in strengthening of the mudstone. Indeed, the UCS doubled when the temperature went from 450 °C to 750 °C.

Future work will include investigations on another sedimentary rock (sandstone) and on the thermal effects on the stress-strain relationships under triaxial conditions as well as tensile strength using Brazillian test. This will provide a more comprehensive dataset on failure criterion of sedimentary rocks under high temperatures.

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