DE GRUYTER OPEN

Acta Geophysica
vol. 64, no. 3, June 2016, pp. 633-648
DOI: 10.1515/acgeo-2016-0021

Variation of Wave Velocity and Porosity of Sandstone after High Temperature Heating

Qiang SUN[1],·Weiqiang ZHANG[1], Tianming SU[2], and Shuyun ZHU[1]

[1]School of Resources and Geosciences,
China University of Mining and Technology, Xuzhou, China
e-mail: sunqiang04@126.com

[2]Research Institute of Highway Ministry of Transport, Beijing, China

A b s t r a c t

This paper reports the variations of mass, porosity, and wave velocity of sandstone after high temperature heating. The range of temperature to which the sandstone specimens have been exposed is 25-850°C, in a heating furnace. It has been shown that below 300°C, porosity and wave velocity change very little. Above 300°C, there is a rapid increase in porosity, but the wave velocity decreases significantly. The results of thermo gravimetric analysis (TGA), differential scanning calorimetry (DSC) and mercury intrusion porosimetry (MIP) suggest that a series of changes occurred between 400 and 600°C in sandstone could be responsible for the different patterns of variation in porosity and wave velocity.

Key words: sandstone, thermal damage, wave velocity, porosity, high temperature treatment.

1. INTRODUCTION

The properties of rocks at high temperature or after heating at high temperature are parameters of growing importance in engineering projects and studies of geological disasters and geological structure formation (Just and Kontny 2012), such as deep geological repositories for heat-generating radioactive wastes (Laloui *et al*. 2008, Zhao *et al*. 2009), exploration of geo-

thermal energy (Zhang *et al*. 2001, Zhang *et al*. 2008, Ranjith *et al*. 2012), deep petroleum boring (Wu *et al*. 2003, Shafiei and Dusseault 2013), underground gasification of coals (Wu *et al*. 2005, Tian *et al*. 2012) and the protection of buildings against fire or building restoration after exposure to fire (Hajpál 2002, Wu *et al*. 2013).

The knowledge on permeability and velocity of wave propagation in rocks under different geological conditions is required in the engineering exploitation of geothermal resource and oil or gas (Somerton *et al*. 1965, Gong and Xie 1989). It is generally accepted that the wave velocity decreases with temperature both under and after high temperature heating treatment (Somerton 1992). However, the porosity increases with temperature (Hajpál 2002) with the increase of porosity, the permeability is also increased gradually (You and Kang 2009).

In most rocks subjected to geological hazards and rock engineering, cracks play an important role, and key information on the structure of cracks in rock mass could be reflected in wave velocity. Variations in the velocity at different temperatures have been considered as a possible precursory phenomenon has been used to evaluate the deformation and fracture feature of rocks (*e.g*., Lebedev and Zharikov 2000, Qin *et al*. 2009, Wu *et al.* 2013, Brantut *et al*. 2013).

In the past few decades, considerable experimental effort has been taken to quantify the relation between wave velocity and cracks (or stress) of rock. Experiments on a variety of rock types involved measurements of typical wave velocities close to rock damage (*e.g*., Rao and Murthy 2001, Nasseri *et al*. 2007, Chaki *et al*. 2008, Lokajíček *et al*. 2012), and the effect of fracture on wave velocity is sufficiently understood. However, the mechanism of wave velocity and porosity variation at high temperature is not clear, probably because the physical and chemical thermal process of rock is not precisely known and the effect of mineral composition is not well understood.

In order to identify the damage level of thermally-cracked sandstone based on the variation of its physical parameters, the variations of some physical properties of sandstone, including mass loss level, porosity, wave velocity, and surface features, were examined after heating treatment in this paper. The heating temperature was varied in a large range, from 25 to 850°C, using a furnace in the laboratory.

2. EXPERIMENTAL TESTS

Sandstone samples were obtained from Linyi, Shandong province. X-ray fluorescence analyzer showed that the oxides incorporated in sandstone include SiO_2, Al_2O_3, K_2O, Na_2O, MgO, CaO, Fe_2O_3, CO_2, *etc.* (Table 1). X-ray diffraction (XRD) analysis showed that the main components are quartz,

Table 1

Chemical composition of sandstone sample

Sample no.	CO_2	MgO	Al_2O_3	SiO_2	K_2O/Na_2O	CaO	Fe_2O_3
1	22.64%	8.10%	10.55	35.49	2.96%	13.43%	5.53%

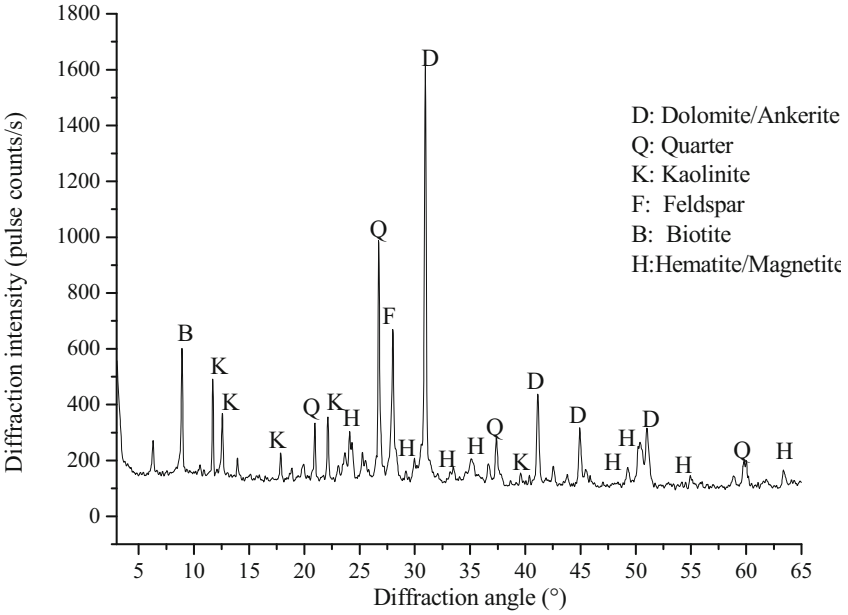


Fig. 1. XRD spectrum of sandstone sample (under 25°C).

feldspar, dolomite/ankerite, and hematite/magnetite structures (Fig. 1), accompanied by a small amount of biotite and kaolinite structure. Therefore, the cement of the sandstone is of ferrigenous, dolomitic, and/or clayey type.

These samples with average bulk density of 2.41 g/cm^3 at room temperature were cut into Φ 50 × 100 mm cylinders. The mass, volume, porosity, and *P*-wave velocity of these specimens were tested before and after heating. The porosity was measured by a microporosity structure analyzer apparatus (type 9310) produced by Micromeritics Instrument Corp., and the *P*-wave velocity was simultaneously collected by a TICO test machine.

The heating apparatus consists of a high temperature furnace (type MTS652.02). The specimens used in all the following tests were placed in the furnace and heated to the designated temperature (25, 75, 100, 150, 200, 250, 300, 330, 340, 350, 400, 450, 500, 550, 570, 585, 600, 700, 800, and

850°C, respectively) at the rate of 30°C/min. Each specimen was kept at its designated temperature for about 30 min before the power was cut off, and the specimen was allowed to cool down naturally with the decline of temperature in the furnace.

The mass, volume, porosity, and *P*-wave velocity of the specimens were tested before and after being heated. The thermo gravimetric analysis (TGA) and differential scanning calorimetry (DSC) of the sandstone specimens were also made with a synchronous comprehensive thermal analyzer (type STA409C) produced by NETZSCH Co., Ltd. In these tests, the heating rate was 5 k/min, and the gas flow rate was 100 mL/min.

3. RESULTS AND DISCUSSIONS

3.1 Variation of surface features

The variations of colour and surface features of the heated sandstone specimens are shown by Fig. 2. From 25 to 400°C, the colour of sandstone faded gradually. Above 400°C, sandstone turns into brick red. Figures 3 and 4 also show that when the heating temperature is higher than 400°C, the colour of sandstone is changed significantly (Su *et al*. 2014). This change resulted from the loss of water of different kind. Free water would escape at around 100°C and the bound water escapes between 100-300°C. Above 300°C, the loss of crystal water and structural water would lead to reduced hydroxyl (-OH), damage of mineral crystal lattice skeleton, increased sandstone defects, and colour change. Additionally, dolomite/ankerite and magnetite, carbonates and organic materials in sandstone are disintegrated into oxides, carbon dioxide, and water when the temperature is above 300°C (especially between 300 to 600°C).

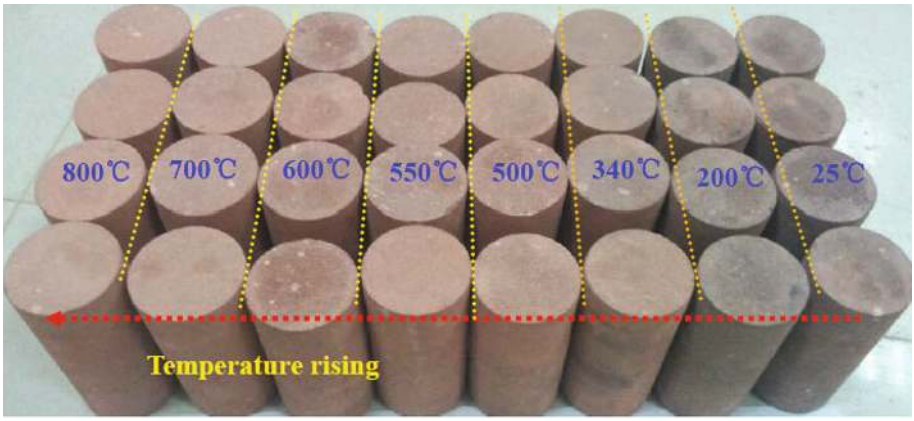


Fig. 2. Color change after different temperature effect for sandstone.

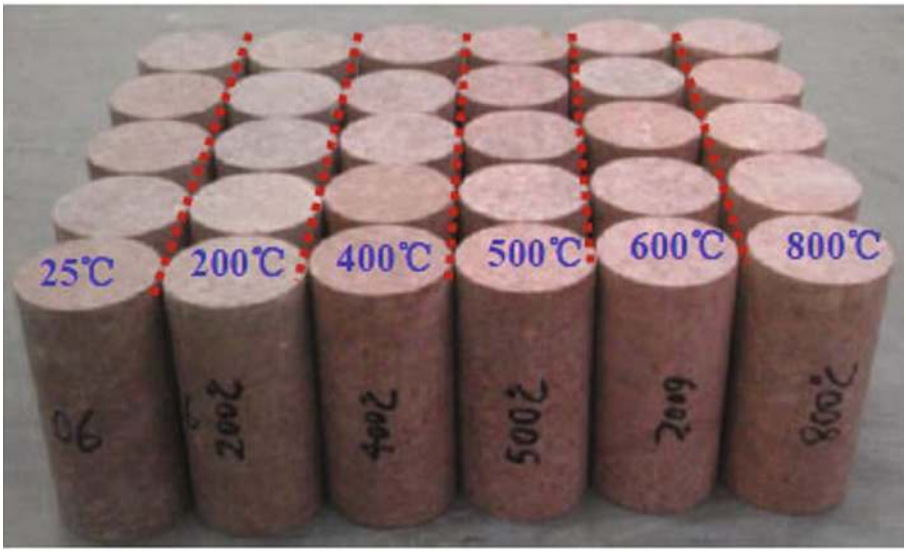
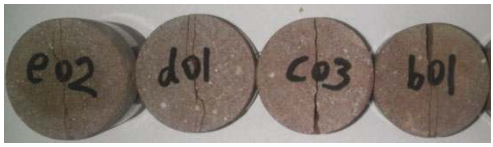


Fig. 3. Sandstone samples after high temperature (Su *et al.* 2014).



(a) 20°C

(b) 400°C

(c) 800°C

Fig. 4. Failure characteristics of Brazilian disc after different treated temperature.

3.2 Variation of mass

The variation of mass loss level *versus* temperature is shown in Fig. 5. Combining the results of TGA and DSC analyses (as shown in Fig. 6), three phases can be identified in the curve shown in Figs. 5 and 7.

(1) Room temperature to 300°C. When the treatment temperature is lower than 300°C, mass loss is generally caused by the evaporation of water. In the process of heating, the state of the water existing inside sandstone would change. The absorbed water, bounded water, and mineral water (*i.e.*, crystal water, structural water, and zeolite water) would escape from sandstone at different temperatures. Free water would escape at around 100°C, and bounded water escapes between 100-300°C.

(2) 300 to 700°C. Above 300°C, the loss of crystal water and structural water would lead to the loss of hydroxyl (-OH), damage of mineral crystal lattice skeleton and increase in the number of defects of sandstone. Additionally, carbonates and organic materials in sandstone are disintegrated into oxides, carbon dioxide, and water when the temperature is above 300°C (especially between 300 to 600°C). At roughly 573°C, quartz experiences a phase transformation from the α phase to β phase (Glover *et al*. 1995); in the

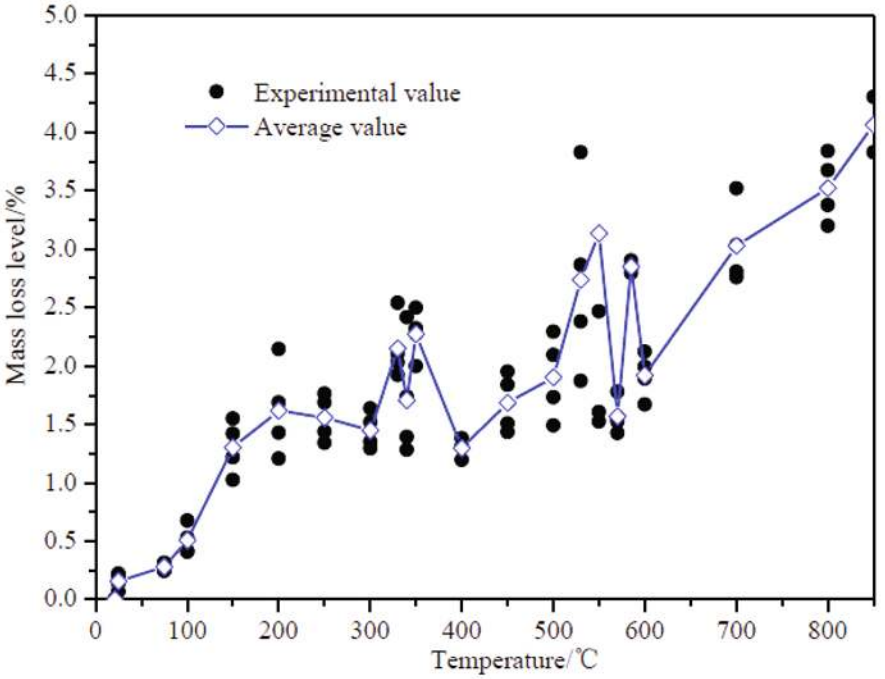


Fig. 5. Variation of mass after different treated temperature.

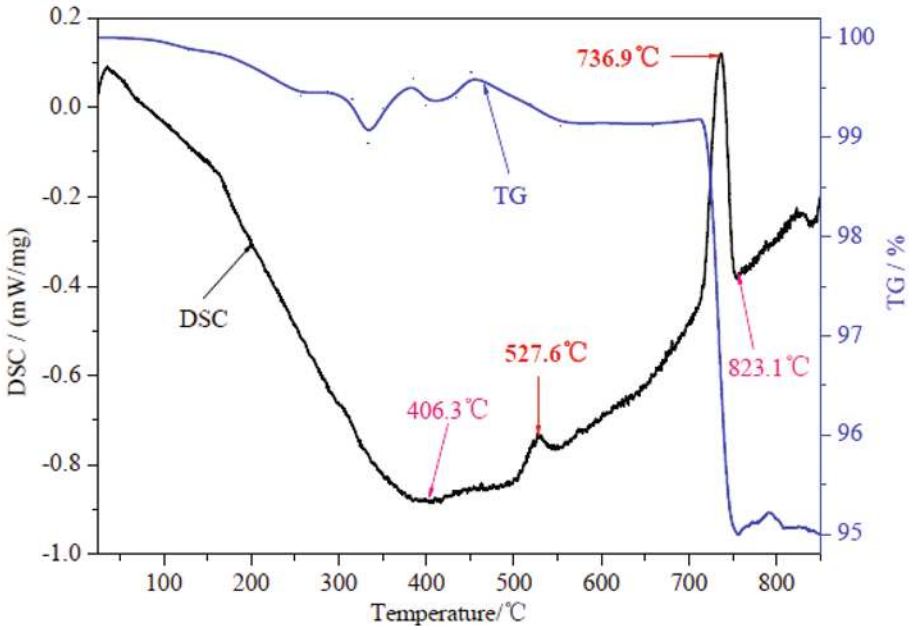


Fig. 6. The curve of DSC-TG for sandstone.

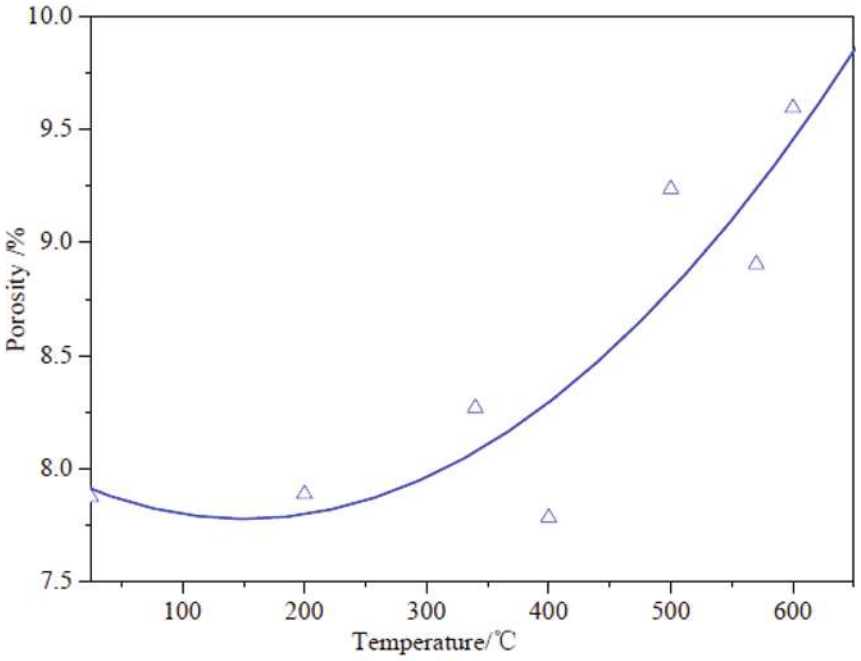


Fig. 7. Variation of porosity after different treated temperature.

range of 470-520°C and above 540°C, dehydration reaction occurs in kaolinte (as shown by Eqs. 1 and 2). Between 400 and 600°C, especially from 500 to 600°C, chemical changes would occur to the minerals such as dolomite/ankerite, magnetite, illite, and kaolinite.

$$Al_2O_3 \bullet 2SiO_2 \bullet 2H_2O \rightarrow Al_2O_3 \bullet 2SiO_2 \bullet \frac{1}{2}H_2O + \frac{3}{2}H_2O \ , \quad (1)$$

$$Al_2O_3 \bullet 2SiO_2 \bullet \frac{1}{2}H_2O \rightarrow Al_2O_3 \bullet 2SiO_2 + \frac{1}{2}H_2O \ , \quad (2)$$

$$2Fe_3O_4 + \frac{1}{2}O_2 \rightarrow 3Fe_2O_3 \ . \quad (3)$$

(3) Over 700°C. When the treatment temperature is higher than 700°C, there is a significant reduction in mass (shown in Fig. 5 and the TG curve in Fig. 6). At roughly 736.9°C, there is an endothermic peak in the DSC curve, corresponding to the change of potassium feldspar into potassium microcline.

3.3 Variation of porosity

The porosity of sandstone after being heated at different temperatures was obtained by Mercury intrusion porosimetry (MIP). The test range of pore diameter is between 0.001 to 200 μm. According to the pore size (pore diameter represented by d), the pores can be divided into four levels: (i) macropore ($d > 100$ μm) – pore liquid can flow freely; (ii) medium-pore ($100 \geq d > 10$ μm) – in the natural state, and when certain head pressure is imposed, pore liquid can flow; (iii) micro-pore ($10 \geq d \geq 1.0$ μm) – under high pressure, the liquid can flow, but the permeability is very low; (iv) smaller micro-pore ($d < 1.0$ μm) – the liquid usually cannot flow.

The variation of total porosity *versus* temperature is shown in Fig. 7. Below 400°C, the porosity changes very little. After that, it increases with temperature. At 600°C, the porosity is nearly 1.23 times the initial value (under 25°C).

Figure 7 also shows a plot of porosity under the atmospheric conditions as a function (shown in Eq. 4) of temperature. The increment of porosity with temperature below 400°C is small, but becomes significant from that temperature onwards. The variation of porosity with temperature may be either caused by thermal expansion and altered micro-crack network, or driven by the structural damage of rocks (Hajpál and Török 2004). The cementing materials and initial porosity of sandstone play very important roles in this process.

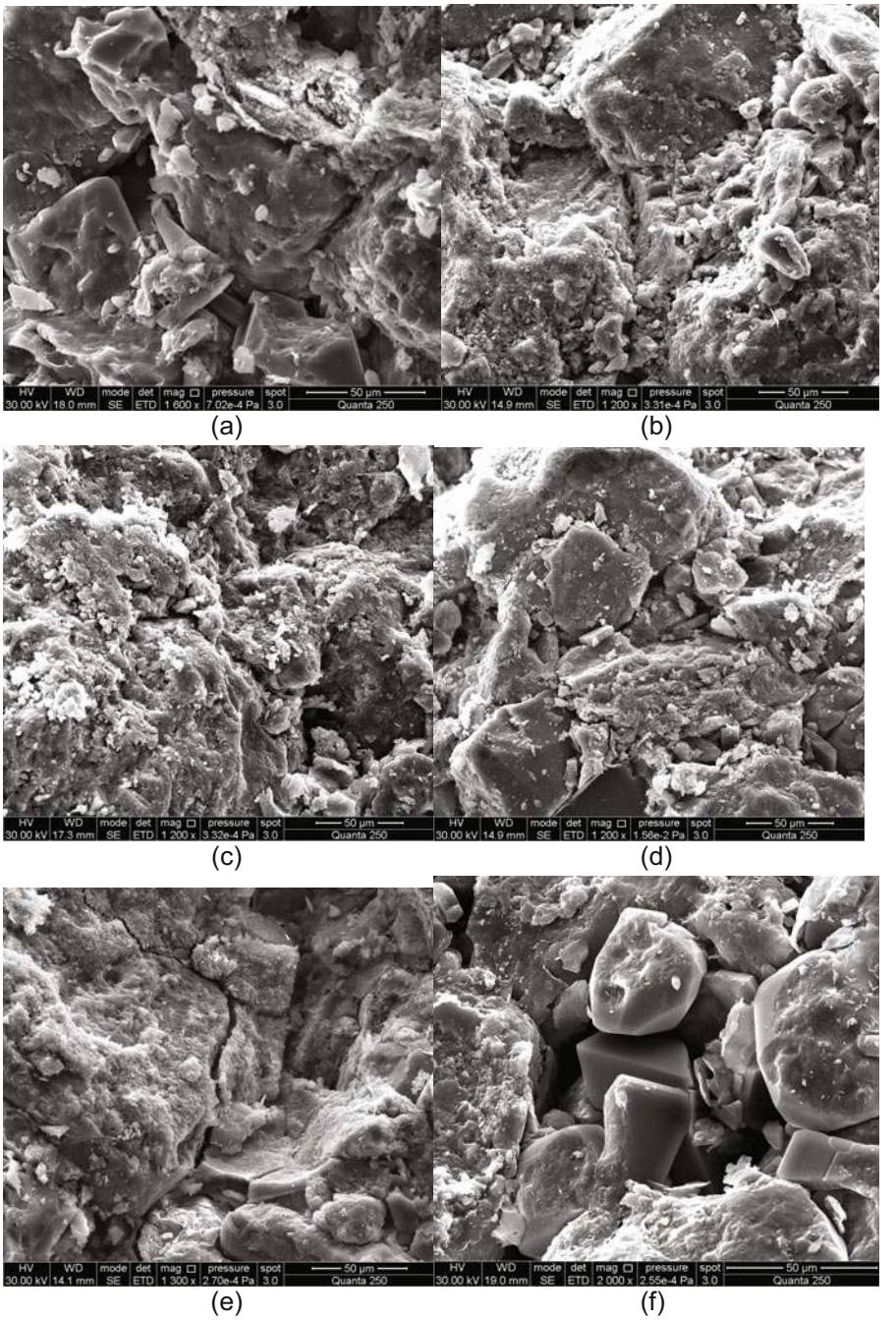


Fig. 8. SEM photographs of sandstone after high temperature (longest side of the photos are 50 μm): (a) 25°C, (b) 200°C, (c) 340°C, (d) 400°C, (e) 570°C, (f) 600°C.

$$n = 7.996677 - 0.00252T + 8.32664 \times 10^{-6} T^2, \quad R^2 = 0.81 , \tag{4}$$

where n is the porosity, and T is the temperature [°C].

Scanning electron microscope analyses were used to record different types of cement materials in the studies of surface grains (Fig. 8). Crystal morphology was used in the identification of mineral phases. The sharp-edged (Fig. 8a and b), opened cracks at minerals contacts (Fig. 8c and d) or even inside of the minerals (Fig. 8e and f) were formed owing to thermal expansion.

Figure 8 shows that with the increasing temperature, the number of microcracks increases significantly, especially at temperatures higher than 400°C, consistent with the MIP result. Additionally, when the temperature is lower, cracks only develop at the edges of grains. When the temperature reaches a threshold, the cemented mineral crystals in sandstone rupture. For example, in Fig. 8f we can find that a large number of fractures have developed on the idiomorphic dolomite cement on quartz grain in the carbonate-cemented sandstone. Porosity increases with the heating temperature; bulk density and wave velocity decrease with the heating temperature.

3.4 Variation of longitudinal wave velocity

The velocity of longitudinal or *P*-wave propagation in the sandstone specimens was examined before and after heating treatment. As wave velocity is closely related to other physical and mechanical properties (density, moisture content, structural characteristics, porosity, *etc*.) of sandstone, the variation of wave velocity due to high temperature may reflect the variation of these properties of sandstone samples. Therefore, determining the change in wave velocity after heating may provide a means of evaluating the effect of heating.

Figure 9 shows that the wave velocity decreases with increasing temperature during heating treatment. The gradient of the curve indicates that there is little variation in wave velocity at temperatures below 300°C, but between 300 and 850°C (especially between 340 and 600°C), there is a significant reduction in wave velocity. Figure 10 shows that the change of wave velocity is correlated to thermal expansion and the variation of micro-crack network, or driven by the structural damage (mass loss level). From the same figure, we may infer that at temperatures higher than 300°C, the temperature effect on wave velocity should be mainly caused by changed mineral content.

In the range of 300 and 600°C, the mineralogical composition and texture of sandstones significantly influence the wave velocity and porosity of sandstone against high temperature. Quartz is the main rock constituent, which shows reversible change at 573°C (α-quartz becomes to β-modifica-

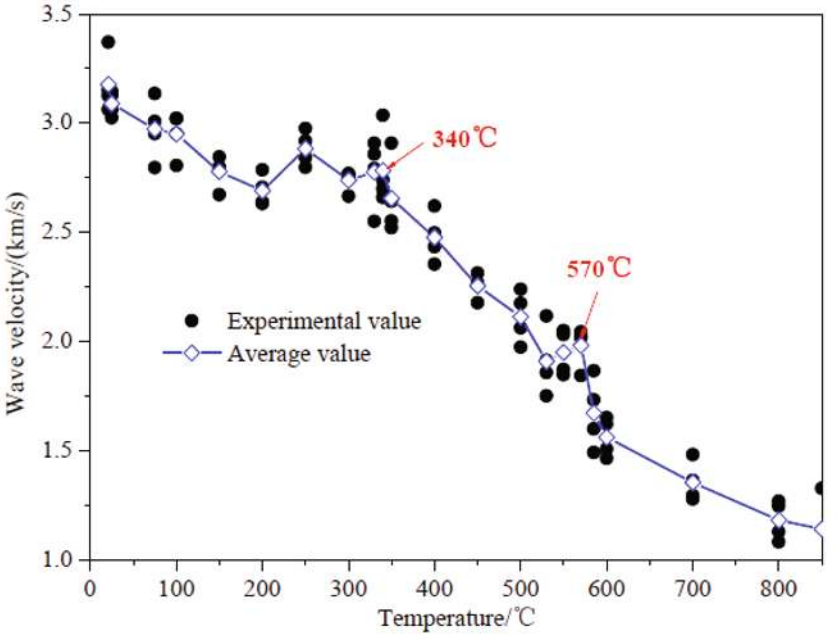


Fig. 9. Variation of wave velocity after different treated temperature.

tion), which causes crack increase. This results in the increase of porosity and decrease of wave velocity. The cemented minerals of dolomite/ankerite and clay or magnetite for sandstone show important changes in mineral composition and in physical properties even at lower temperatures. The most important change is that the structure of clay minerals and carbonate disintegrates when the heating temperature is higher than 450°C. Additionally, above 300°C, the loss of crystal water and structural water would lead to the loss of hydroxyl (-OH), damage of mineral crystal lattice skeleton, and increase the number of defects of sandstone. After high temperature heating, sandstone can be seriously damaged. With increasing damage, the wave velocity decreases quickly.

According to the analysis presented above, the thermal damage of sandstone gets larger as the temperature increases. The relation between wave velocity and damage factor can be described by the equation (Zhao *et al.* 2009).

$$D \approx 1 - \left(\frac{V_p}{V_0} \right)^2 , \tag{5}$$

where D is the damage index, V_0 is the initial wave velocity (before heating), and V_p is the wave velocity after heating.

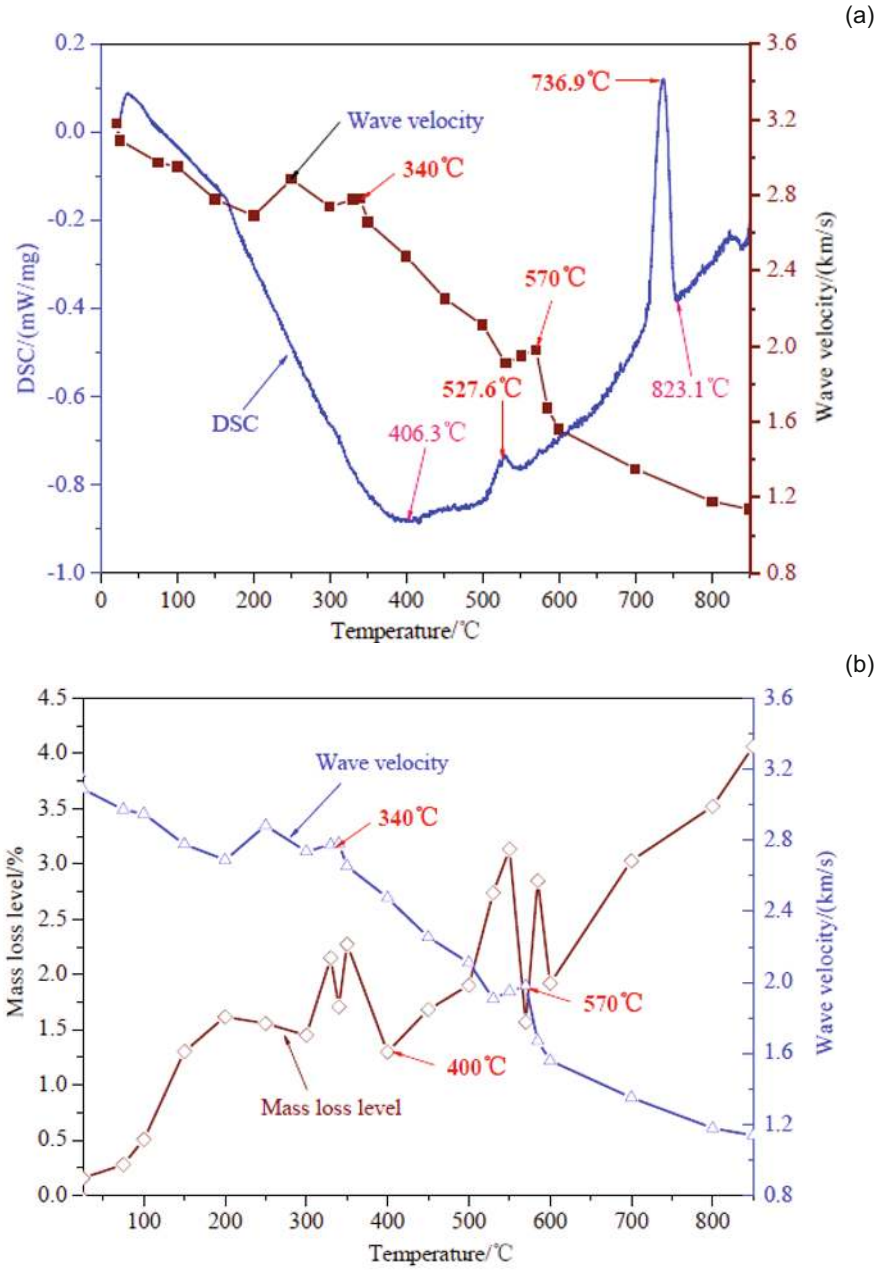


Fig. 10. Variation of wave velocity after different treated temperature: (a) relationship between wave velocity, DSC, and temperature; (b) relationship between wave velocity, mass loss level, and temperature.

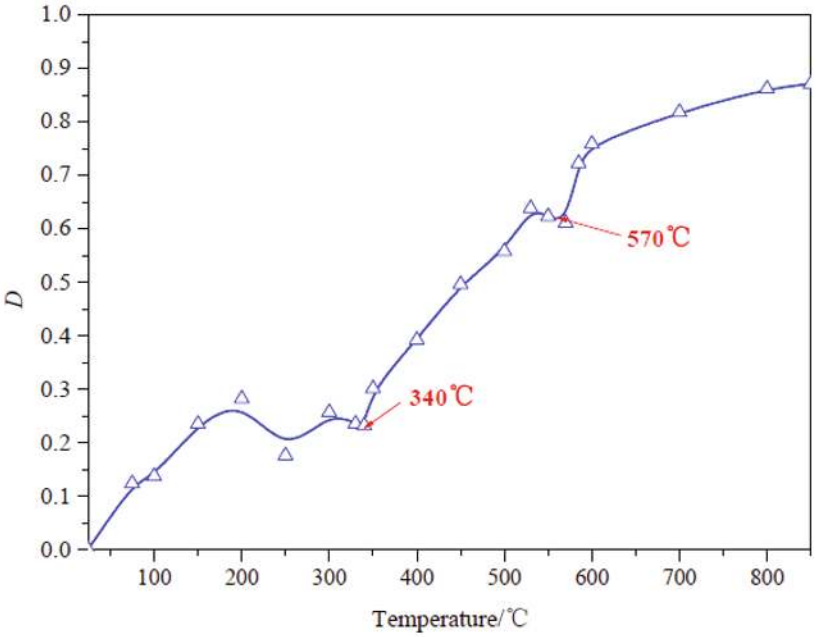


Fig. 11. Variation damage index after different treated temperature.

The measured temperature effect on thermal damage, obtained in this study, is shown in Fig. 11, which clearly displays that above 340°C, the thermal damage of sandstone increases rapidly with temperature.

4. CONCLUSION

To assess the effect of high temperature on sandstone, the characteristics of mass, pores, wave velocity, and surface features of sandstone specimens before and after heating at different temperatures have been examined in this paper. The major findings in this study are listed below:

- Being heated at temperatures from 25 to 300°C, the sandstone color faded gradually. Above 400°C, sandstone turns into brick red.
- The increase in porosity depends on the mineralogy of particles and cement. The minor cracks, quartz, feldspars and dolomite/ankerite, agnetite and clay minerals show significant changes. Sandstone mineral cements often show changes in mineral composition or they are even destroyed at high temperatures. The most important change is that the structures of minerals disintegrate between 400 and 600°C.
- Between 300 and 850°C (especially between 340 and 600°C), there is a significant reduction in wave velocity. The change of wave velocity is correlated to structural damage and the variation of mineral content.

Acknowledgments. This research was supported by the State Basic Research and Development Program of China (No. 2013CB036003), the Priority Academic Program Development of Jiangsu Higher Education Institutions, Transport project (2013318J12330), and the National Science Youth Foundation of China (Grant No. 41102201, No. 41302233).

References

Brantut, N., M.J. Heap, P.G. Meredith, and P. Baud (2013), Time-dependent cracking and brittle creep in crustal rocks: A review, *J. Struct. Geol*. **52**, 17-43, DOI: 10.1016/j.jsg.2013.03.007.

Chaki, S., M. Takarli, and W.P. Agbodjan (2008), Influence of thermal damage on physical properties of a granite rock: porosity, permeability and ultrasonic wave evolutions, *Constr. Build. Mater*. **22**, 7, 1456-1461, DOI: 10.1016/j.conbuildmat.2007.04.002.

Glover, P.W.J., P. Baud, M. Darot, P.G. Meredith, S.A. Boon, M. LeRavalec, S. Zoussi, and T. Reuschlé (1995), α/β phase transition in quartz monitored using acoustic emissions, *Geophys J Int*. **120**, 3, 775-782, DOI: 10.1111/j.1365-246X.1995.tb01852.x.

Gong, Y.G., and Y.D. Xie (1989), A review of laboratory investigation of rock permeability, *Chin J. Rock Mech. Eng.* **8**, 3, 219-227, DOI: 10.3321/j.issn:1000-6915.1989.03.001 (in Chinese).

Hajpál, M. (2002), Changes in sandstones of historical monuments exposed to fire or high temperature, *Fire Technol.* **38**, 4, 373-382, DOI: 10.1023/A:1020174500861.

Hajpál, M., and Á. Török (2004), Mineralogical and color changes of quartz sandstones by heat, *Environ. Geol*. **46**, 3, 311-322, DOI: 10.1007/s00254-004-1034-z.

Just, J., and A. Kontny (2012), Thermally induced alterations of minerals during measurements of the temperature dependence of magnetic susceptibility: a case study from the hydrothermally altered Soultz-sous-Forêts granite, France, *Int. J Earth Sci*. **101**, 3, 819-839, DOI: 10.1007/s00531-011-0668-9.

Laloui, L., B. François, M. Nuth, H. Peron, and A. Koliji (2008), A thermo-hydro-mechanical stress-strain framework for modelling the performance of clay barriers in deep geological repositories for radioactive waste. **In:** D.G. Toll, C.E. Augarde, D. Gallipoli, and S.J. Wheeler (eds.), *Unsaturated Soils: Advances in Geo-Engineering*, Taylor & Francis Group, London, 63-80.

Lebedev, E.B., and A.V. Zharikov (2000), Study of intergranular films and interstitial phases in geomaterials using high temperature centrifuge and ultrasonic method at high pressure, *Phys. Chem. Earth A*, **25**, 2, 209-214, DOI: 10.1016/S1464-1895(00)00033-8.

Lokajíček, T., V. Rudajev, R.D. Dwivedi, R.K. Goel, and A. Swarup (2012), Influence of thermal heating on elastic wave velocities in granulite, *Int J. Rock Mech. Min. Sci.* **54**, 1-8, DOI: 10.1016/j.ijrmms.2012.05.012.

Nasseri, M.H.B., A. Schubnel, and R.P. Young (2007), Coupled evolutions of fracture toughness and elastic wave velocities at high crack density in thermally treated Westerly granite, *Int. J. Rock Mech. Min. Sci.* **44**, 4, 601-616, DOI: 10.1016/j.ijrmms.2006.09.008.

Qin, B.D., J. He, and L.J. Chen (2009), Experimental research on mechanical properties of limestone and sandstone under high temperature, *J. Geomech*. **15**, 3, 253-261, DOI: 10.3969/j.issn.1006-6616.2009.03.007 (in Chinese).

Ranjith, P.G., D.R. Viete, B.J. Chen, and M.S.A. Perera (2012), Transformation plasticity and the effect of temperature on the mechanical behavior of Hawkesbury sandstone at atmospheric pressure, *Eng. Geol*. **151**, 120-127, DOI: 10.1016/j.enggeo.2012.09.007.

Rao, G.M.N., and C.R.L. Murthy (2001), Dual role of microcracks: toughening and degradation, *Can. Geotech. J.* **38**, 2, 427-440, DOI: 10.1139/t00-105.

Shafiei, A., and M.B. Dusseault (2013), Geomechanics of thermal viscous oil production in sandstones, *J. Petrol. Sci. Eng.* **103**, 121-139, DOI: 10.1016/j.petrol.2013.02.001.

Somerton, W.H. (1992), *Thermal Properties and Temperature-Related Behavior of Rock/Fluid Systems*, Developments in Petroleum Science, Vol. 37, Elsevier, Amsterdam.

Somerton, W.H., M.M. Mehta, and G.W. Dean (1965), Thermal alteration of sandstones, *J. Petrol. Technol.* **17**, 5, 589-593, DOI: 10.2118/907-PA.

Su, H.J., H.W. Jing, and H.H. Zhao (2014), Experimental investigation on loading rate effect of sandstone after high temperature under uniaxial compression, *Chin. J. Geotech. Eng*. **36**, 6, 1064-1071, DOI: 10.11779/CJGE201406011 (in Chinese).

Tian, H., T. Kempka, N.X. Xu, and M. Ziegler (2012), Physical properties of sandstones after high temperature treatment, *Rock Mech. Rock Eng.* **45**, 6, 1113-1117, DOI: 10.1007/s00603-012-0228-z (in Chinese).

Wu, G., Y. Wang, G. Swift, and J. Chen (2013), Laboratory investigation of the effects of temperature on the mechanical properties of sandstone, *Geotech. Geol. Eng*. **31**, 2, 809-816, DOI: 10.1007/s10706-013-9614-x.

Wu, X.D., J.R. Liu, and J.S. Qin (2003), Effects of thermal treatment on wave velocity as well as porosity and permeability of rock, *J. Univ. Petrol. China* **27**, 4, 70-72, DOI: 10.3321/j.issn:1000-5870.2003.04.018 (in Chinese).

Wu, Z., B.D. Qin, L.J. Chen, and Y.J. Luo (2005), Experimental study on mechanical character of sandstone of the upper plank of coal bed under high temperature, *Chin. J. Rock Mech. Eng*. **24**, 11, 1863-1867, DOI: 10.3321/j.issn:1000-6915.2005.11.008 (in Chinese).

You, L.J., and Y.L. Kang (2009), Effects of thermal treatment on physical property of tight rocks, *Prog. Geophys.* **24**, 5, 1850-1854, DOI: 10.3969/j.issn.1004-2903.2009.05.040.

Zhang, Y., Y.S. Zhao, Z.J. Wan, F. Qu, F.K. Dong, and Z.J. Feng (2008), Experimental study on effect pore pressure on feldspar fine sandstone permeability under different temperatures, *Chin. J. Rock Mech. Eng.* **27**, 1, 53-58, DOI: 10.3321/j.issn:1000-6915.2008.01.008 (in Chinese).

Zhang, Z.X., J. Yu, S.Q. Kou, and P.-A. Lindqvist (2001), Effects of high temperatures on dynamic rock fracture, *Int. J. Rock. Mech. Min. Sci*. **38**, 2, 211-225, DOI: 10.1016/S1365-1609(00)00071-X.

Zhao, H.B., G.Z. Yin, and L.J. Chen (2009), Experimental study on effect of temperature on sandstone damage, *Chin. J. Rock Mech. Eng*. **28**, S1, 2784-2788, DOI: 10.3321/j.issn:1000-6915.2009.z1.030 (in Chinese).

Received 25 November 2014
Received in revised form 21 April 2015
Accepted 10 May 2015