RESEARCH ARTICLE

Influence of sintering temperature on the characteristics of shale brick containing oil well-derived drilling waste

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Abstract

Introduction The influence of sintering temperature on the physico-mechanical characteristics (such as water absorption, apparent porosity, bulk density, weight loss on ignition, firing shrinkage, and compressive strength), leachability, and microstructure of shale brick containing oil well-derived drilling waste (DW) was investigated.

Methods The experiments were conducted at a temperature ranging from 950°C to 1,050°C with 30% DW addition.

Results The results indicate that increasing the sintering temperature decreases the water absorption and apparent porosity and increases the shrinkage, density, and compressive strength of sintered specimens. Moreover, the physico-mechanical properties of samples sintered at 1,050°C meet the requirements of the MU20 according to GB/T 5101-2003 (in China). The heavy metal concentrations of the leachate are much lower than the current regulatory limits according to GB16889-2008.

Conclusion The results from XRD and SEM show that increasing sintering temperature results in an increase of the high temperature liquid phase, which may have a significant effect on the densification process of the samples.

Keywords Oil well-derived drilling waste (DW) · Shale brick · Sintering temperature · Physico-mechanical property · Leachability

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1 Introduction

In the petroleum industry, various types of waste materials are generated including oil well-derived drilling waste (DW), which contains significant concentrations of heavy metals and organic pollutants as well as others, making necessary treatment and confined disposal to manage them properly. Therefore, the petroleum industry is confronted with the disposal problem of the huge amount of accumulated waste materials produced over the years. At present, the main predominant waste disposal methods include landfilling, sealing in the pit, incinerating, microbiological processing, mud to cement, and stabilization/solidification (S/S) (Peng et al. 2007; Yang et al. 2003; Charles et al. 2006; Xu et al. 2006; Tuncan et al. 1997). But in the long run, harmful substances will leak to the outside world, which still will bring pollution to the environment.

Sintering treatment technology is becoming more attractive as an alternative method for the recovery and recycling of inorganic wastes and residues. The prospective benefits of using wastes and residues in the production of sintered brick include immobilizing heavy metals in the fired matrix, oxidizing organic matter, and destroying any pathogens during the firing process. So many researchers have successfully developed suitable products (such as lightweight aggregate, bricks, tiles, and other construction products) made from various wastes or residues by sintering such as reservoir sediment, municipal sludge, bottom ash, fly ash, red mud, reservoir sediment, etc. (Lin et al. 2005, 2006; Mangialardi 2001; Lin 2006; Zhang et al. 2007; Chiang et al. 2008; Weng et al. 2003; Taner 2006). At present, most domestic and overseas researches on oil well-derived drilling waste stress on detoxification treatment, but its resource is not utilized. Although, the recycling of oil well muds for the production of bricks has been recently reported

(El-Mahllawy and Osman 2010), they used clay as the main material, which is one kind of key and decreasing resources. Actually, the Chinese government has already enacted a law to prohibit using clay to fabricate sintered solid clay brick. So, in this paper, shale was used as the main material instead of clay, and there is rarely research on the utilization of oil well-derived waste in the production of shale brick. This study will potentially provide further beneficial uses of oil well-derived waste.

Therefore, in the present paper, the feasibility of oil well-derived drilling waste used as a replacement material for producing shale brick was investigated. In addition, relationships among sintering temperature, microstructure, and characteristics of sintered shale brick production containing oil well-derived drilling waste were further discussed. For this purpose, experiments were conducted on physico-mechanical properties (water absorption, apparent porosity, bulk density, weight loss on ignition, firing shrinkage) and mechanical property (compressive strength) of sintered specimens. Mineralogy and microstructure of sintered specimens were also discussed by X-ray diffraction (XRD) and scanning electron microscopy (SEM) analysis, respectively.

2 Experiments

2.1 Raw materials

The oil well-derived drilling waste and shale used in this study were collected from Sichuan and Qinhuangdao in China, respectively. Prior to further applications, both the raw materials were dried and ground into fine powder until the particles could pass through a 75-μm mesh sieve. The specific surface area of DW and shale are 486 and 618 m^2/kg, respectively; their particle size distributions determined by Mastersizer 2000E are shown in Fig. 1. The physical properties and Atterberg limits of raw materials are summarized in Tables 1 and 2, respectively. The chemical composition of the raw materials, as analyzed by XRF, is shown in Table 3.

As shown in Table 3, the major components in the shale are SiO_2 (58.89%), Al_2O_3 (18.75%), and Fe_2O_3 (8.95%). DW has a high amount of BaO (30.04%) and SiO_2 (58.89%) as well as CaO (18.24%), which will cause lime burst in the sintered bricks. Besides, a higher loss on ignition of DW is also not favored for the production of sintered bricks; this is related to the development of the porosity and the densification in the process of sintering and eventually has an effect on the compressive strength of the sintered products (Lin et al. 2005; Weng et al. 2003). Consequently, DW was mixed in the raw material at a relatively low proportion (30%) in this study.
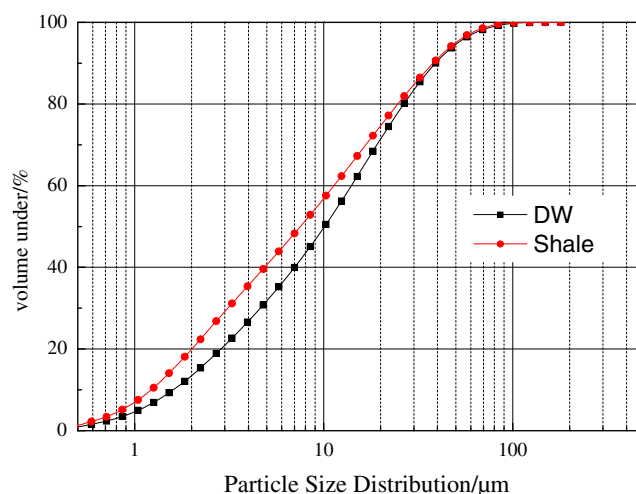

Fig. 1 Particle size distribution of DW and shale

2.2 Sample preparation

DW and shale were homogenized in a blender by the proportion of 30:70 (by weight), and the mixtures were cured with the addition of 15% water (by weight) in a humidity chamber with a temperature of 20±1°C and a relative humidity of 98±2% for 72 h. Cylindrical green samples were prepared by semi-dry pressing method at 20 MPa for 30 s with the size of φ50×40 mm. Green samples were dried at room conditions (25°C, 40% relative humidity) for 48 h and then oven dried at 105°C for 24 h to drive off the moisture prior to the sintering process.

The dried samples were sintered in an air-ventilated electrical furnace according to the following designed heating program. Heating rate was 2°C/min below 400°C, then 3°C/min from 400°C to the sintering temperature, and kept at sintering temperature for 1 h. Sintered specimens were cooled down to room temperature in the furnace. For each species, six samples were prepared for the following tests, and averages and the coefficient of variation were computed to provide statistics and error estimates.

2.3 Testing methods

Chemical and thermal analyses of raw materials were performed by XRF (Axios-Advanced) and TG/DSC (NETZSCH SAT 449C) techniques, respectively. Mineralogical analyses of raw materials and sintered specimens were

Table 1 Characteristics of DW and shale

Characteristics	Density/(g cm^{-3})	Moisture content/(%)
DW	2.83	8.48
Shale	2.37	5.71

Table 2 Atterberg limits of DW, shale, and mixtures

	DW	Shale	30% DW+70% shale
Liquid limit (%)	40.99	31.02	33.87
Plastic limit (%)	29.75	15.87	21.54
Plasticity index	11.24	15.15	12.33

investigated by X-ray diffraction (XRD-Rigaku D/Max–RB) using Cu-Kα radiation. Microstructure of sintered specimens was investigated by SEM. The particle size distribution of raw materials was determined by Mastersizer 2000E.

Water absorption, apparent porosity, and bulk density of samples soaked in distilled water for 24 h were tested using Shimadzu Balances (AUW220H) basing on the Archimedes principle (Wu 2002). Firing shrinkage was measured by a micrometer. Weight loss on ignition was measured by an electronic analytical balance (TIANYI FA2004). Compressive strength tests were performed using a compression testing machine (TYE-300) at a loading rate of 0.6 kN/s.

Heavy metal leachability experiment was carried out according to the solid waste-extraction procedure for leaching toxicity-acetic acid buffer solution method (HJ/T 300–2007) with samples damaged less than 5 mm, and the leaching solution was analyzed by inductively coupled plasma atomic-emission spectroscopy.

3 Results and discussion

3.1 Thermal analysis

In order to decide the appropriate sintering temperature of shale brick containing DW, the thermal analyses of DW and shale were investigated by TG/DSC. The results of TG/DSC are shown in Fig. 2.

The TG/DSC curves of DW (Fig. 2a) can be explained as follows: (1) An endothermic peak occurs at 82°C due to dehydration (loss of free moisture); (2) Two exothermic peaks occur at 335°C and 454°C with a mass loss due to the combustion of oil and other organic compounds; (3) A second endothermic peak, due to the decomposition of calcite occurs at 710°C; (4) Two endothermic peaks occur at about 1,170°C and 1,210°C with a mass loss of 9.5% due to the decomposition of barite (Ren and Feng 1986).

As shown in Fig. 2b, no obvious endothermic or exothermic peak occurs during the heating process, and the total mass loss was only 3.6%; the DSC curve becomes steep after 1,000°C, and the TG curve shows that there is no quality change; it means the emergence of a large amount of high temperature liquid phases, which can enhance the connection between particles of raw materials and are favorable for the melting of phases and reacting of
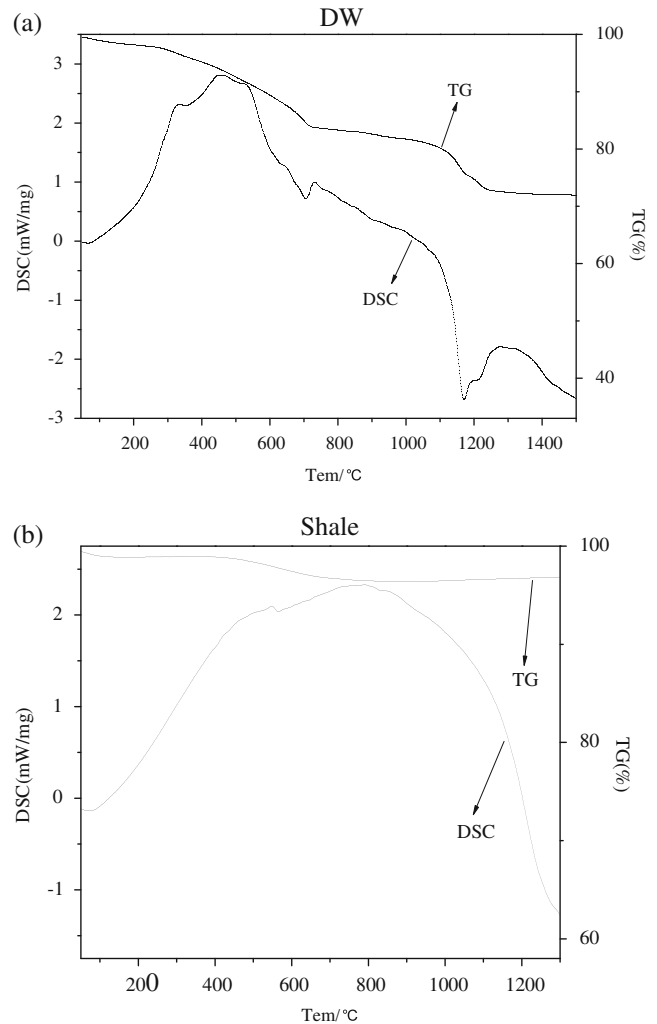
Table 3 Chemical composition of DW and shale (%wt)

Oxide	DW	Shale
SiO$_2$	26.19	58.89
Fe$_2$O$_3$	3.56	8.95
Al$_2$O$_3$	–[a]	18.75
CaO	18.24	0.31
MgO	1.11	2.50
K$_2$O+Na$_2$O	1.10	5.26
BaO	30.64	–[a]
SO$_3$	2.14	0.01
P$_2$O$_5$	0.03	0.09
MnO	0.09	0.08
TiO$_2$	0.35	0.94
SrO	0.28	–[a]
Cl	0.07	–[a]
I.L.	16.11	4.20

[a] <0.01%

Fig. 2 Thermal analysis of raw materials

Table 4 Physico-mechanical properties of the samples sintered at different temperatures

Sintering temperature (°C)	Water absorption (%)	Apparent porosity (%)	Bulk density (g/cm³)	Sintering shrinkage (%)	Weight loss on ignition (%)	Compressive strength (MPa)
950	21.07	36.86	1.73	0.27	8.80	13.79
1,000	20.39	35.74	1.75	0.61	9.10	19.87
1,050	18.27	33.00	1.80	1.15	9.54	24.65

components. According to the above analysis, three sinter-ing temperatures (950°C, 1,000°C, and 1,050°C) were selected.

3.2 Physico-mechanical properties

The effects of sintering temperature on physical properties (such as water absorption, apparent porosity, bulk density, firing shrinkage, and weight loss on ignition) and compressive strength are shown in Table 4.

Water absorption is an effective index in evaluating the quality and densification of building brick. The less water infiltrates into the brick, the more the durability of the brick and resistance to the natural environment are expected. As shown in Table 4, the water absorption of sintered speci-mens decreases from 21.07% to 18.27% with the increasing of the sintering temperature from 950°C to 1,050°C, which is due to the apparent porosity of sintered specimens which decreases significantly with increasing sintering temperature similar to the results accepted by Lin Kae Long (Lin 2006). The smaller water absorption that occurred after sintering at the higher temperature (1,050°C) suggests that high temper-ature liquid phases occurred, which contributed to a decrease in the pore volume and thus the water absorption.

According to the results of bulk density, the bulk density increases from 1.67 to 2.00 gcm^{-3} with increasing temperature, which is closely related to the results of water absorption and apparent porosity. When the sample absorbs more water, the sample exhibits a higher apparent porosity, resulting in a light density.

The dimensional change in sintered specimens is a prominent indicator for the building brick quality. Table 4 reveals that the shrinkage rate increases significantly with the increasing of sintering temperature. The sintered speci-mens produce a significant densification, resulting in total

Table 5 Heavy metal leachability of the samples sintered at different temperatures (milligrams per liter)

Heavy metal	950°C	1,000°C	1,050°C	GB16889-2008
Cr	0.063	0.025	–[a]	1.5
Zn	0.574	0.187	0.073	100

[a] Detection limits<0.007 mg/L

volume shrinkage. Accordingly, a contrary trend occurs between the dimensional change and water absorption of sintered specimens (as shown in Table 4). It can be concluded that the better conditions for sintered specimens are higher shrinkage rate and lower water absorption.

The compressive strength is the most important engi-neering quality index for building materials, and the results indicate that the compressive strength is mainly affected by the sintering temperature. The sintering temperature increases the compressive strength through densification. With the increase of sintering temperature, the compressive strength of samples increases from 13.79 to 24.65 MPa. And sintered at 1,050°C, the strength measured of the samples meets the requirements of the MU20 according to the Chinese National Standard (GB/T 5101–2003 2003). The results indicate that the optimum sintering temperature for maximum compressive strength was 1,050°C, and oil well-derived drilling waste can be used as a replacement material for the production of shale brick.

All the above results indicate that sintering temperature is a key factor determining the brick quality, and compres-sive strength is closely related to the physical properties.
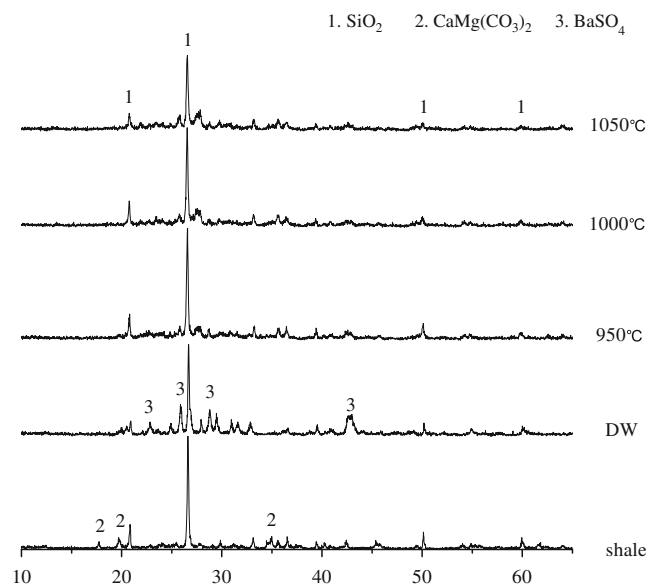
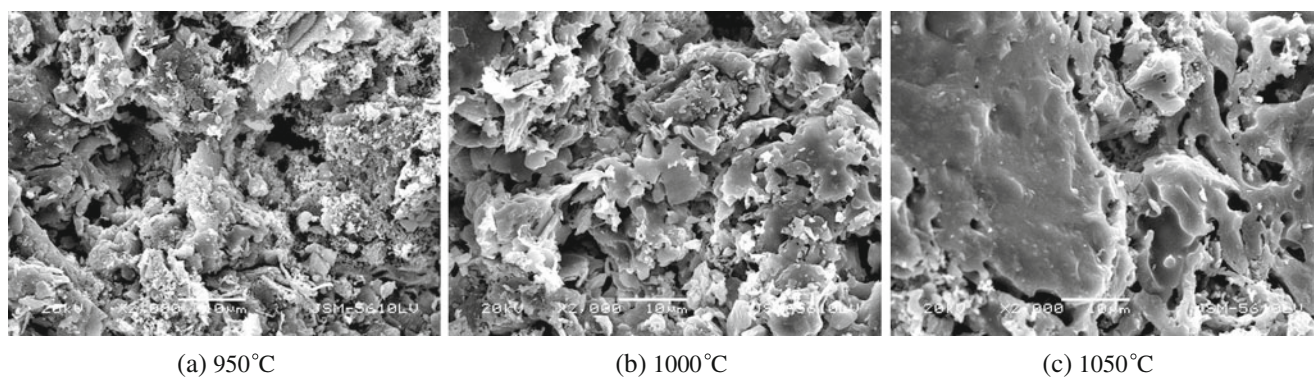**Fig. 3** XRD patterns of raw materials and sintered specimens

(a) 950°C   (b) 1000°C   (c) 1050°C

Fig. 4 SEM images of specimens sintered at different temperatures (×2,000)

3.3 Heavy metal leachability

In the process of oil and gas drilling, some additives contain a lot of heavy metals such as chrome and zinc, bringing a large amount of Cr and Zn in the drilling mud. So, in this paper, a test of heavy metal leachability was carried out only on the heavy metals Cr and Zn, and the results of the specimens sintered at different temperatures are summarized in Table 5.

As shown in Table 5, the total leachate concentrations for all tested metals (Cr and Zn) in the sintered specimens reveal a decreasing tendency with the increase of the sintering temperatures, and the leachate concentrations for the tested metals in all sintered specimens are much lower than the permission range of standard value according to the Chinese National Standard (GB16889-2008). In the process of high temperature sintering, nonvolatile species become chemically bonded into the resultant matrix, rendering them nonleachable (Cortez et al. 1996). So, heat treatment has been identified as a potentially effective tool for immobilizing heavy metals into a nonleachable slag.

Actually, compared with the experimental condition, sintered specimens leaching in situ is a slow and gradually diluting process, and only few grains are below 5 mm, so the heavy metal immobilizing of sintered specimens could be more reliable in the practical environment, and a minimum of harmful materials can be contained and released in the environment.

3.4 XRD analysis

Figure 3 shows the X-ray diffraction pattern of raw materials and the samples sintered at different temperatures (950°C, 1,000°C, 1,050°C). The major mineral phases are quartz (SiO_2) and dolomite ($CaMg(CO_3)_2$) in shale. The DW is mainly composed of quartz (SiO_2) and barite ($BaSO_4$). XRD result comparisons for the as-received sediment samples and sintered specimens at different sintering temperatures indicate that the mineral species

peaks of dolomite and barite disappeared due to their decomposition in the sintering process, which is in agreement with the result of weight loss on ignition(as shown in Table 2); the major mineral phases of the sintered specimens are quartz, and the intensities of quartz peak decreased with sintering temperature increased from 950°C to 1,050°C, which indicated that emergence of increasing amount of high temperature liquid phases, which are proved in the results of physico-chemical and mechanical properties.

3.5 SEM investigation

SEM investigations were conducted in order to get a better understanding of the morphology of the microstructure and the relationships between physico-mechanical properties and microstructure of the sintered specimens. The SEM results of the sintered specimens at different sintering temperatures are illustrated in Fig. 4.

It is clear that the high temperature liquid phases in sintered specimens, which are favorable for the melting and reacting of raw material components, increased in accordance with increasing sintering temperatures. Furthermore, the particles of sintered specimens sintered at 1,050°C are very closely connected together and have a relatively densified structure. That is, the densified matrix material sintered at 1,050°C shows better surface characteristics than that sintered at 1,000°C (as shown in Fig. 4). As previous results indicate, after being sintered at 1,050°C, the sintered specimens have a good quality, higher compressive

Table 6 The properties of standard fired bricks

External appearance	No cracking
Compressive strength (MPa)	18.6
Density (kg/m³)	1,780
Water absorption (%)	19.74
Cracking by lime	No
Frost	No

strength, and lower leaching toxicity of the building brick than the specimens sintered at 950°C and 1,000°C.

3.6 Properties of standard fired common bricks

Standard fired common bricks were prepared with a size of 240×115×53 mm according to the Chinese National Standard (GB5101-2003) with the proportion of DW/shale=30:70 by weight, and fired at 1,000°C. The properties of fired common bricks were listed in Table 6.

4 Conclusion

This study has demonstrated a feasible way of using oil well-derived drilling waste as a partial substitution for shale to produce good quality bricks. Sintered samples were heated to sintering temperatures varying from 950 to 1,050°C. Based on the experiment reported in this paper, the following conclusions and recommendations are drawn:

1. The sintered temperature is a key factor determining the brick quality and compressive strength. Water absorption and apparent porosity decrease with the increase of the sintering temperature, while bulk density and firing shrinkage increase. Compressive strength of the samples increases with the increase of sintering temperature. The physico-mechanical properties of samples sintered at 1,050°C meet the requirements of the MU20 according to the Chinese National Standard (GB/T 5101–2003 2003).
2. Increasing sintering temperature results in an increase of the high temperature liquid phase, which may be have a significant effect on the densification process of the samples.
3. The leachate concentrations of Cr and Zn in all sintered specimens are lower and also in compliance with the permission range of standard value according to the Chinese National Standard (GB16889-2008 2008).

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