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Energy Procedia 158 (2019) 1712–1717
Energy Procedia
www.elsevier.com/locate/procedia

10th International Conference on Applied Energy (ICAE2018), 22-25 August 2018, Hong Kong, China

Effects of Microwave-enhanced Pretreatment on Oil Shale Milling Performance

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Abstract

Oil shale, as an unconventional fossil fuel, exhibits unique properties compared with coal and other petroleum. Due to the nature of sedimentary rock, large amounts of inorganic mineral impurities in rock matrix reduce the grade of oil shale, whilst increase the grinding resistance. This investigation presents the effects of microwave-enhanced pretreatment on the nature of oil shale and compared with conventional preheating process. Two Chinese oil shale from Fushun and Xingsheng Deposits were grounded and sieved into a size fraction (1-1.18mm) and were cut into eighteen cube-shaped specimens respectively. The prepared samples were processed accordingly to investigate how the grindability changed, in comparison to that of raw samples, and how the fundamental chemical properties of oil shale were altered after pretreatment. Quantitive data were used to assess the effects of different pretreatment methods on oil shale milling performance in a lab-scale pulverizer along with the impacts on moisture content, chemical properties. The uniaxial compressive strength (σ_{max}) of Fushun oil shale was reduced 63.1% and the breakage rate increased 44.9% by short exposure to microwave irradiation. In conclusion, microwave-enhanced pretreatment presents significant improvement in oil shale milling performance compared to conventional preheating process in terms of breakage rate (S_i) and uniaxial compressive strength (σ_{max}) which showed negligible alterations.

Keywords: Oil shale grindability; Microwave heating; Uniaxial compressive strengh; Breakage model;

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1876-6102
10.1016/j.egypro.2019.01.398

1. Introduction

The increasing demand of clean liquid fuels has facilitated the revival of unconventional petroleum resources in the new policies Scenarios[1]. Oil shale, a fine-grained sedimentary rock that contains desirable amount of organic matter in its rock matrix, can be converted to crude oil by thermal degradation [2] [3]. Among the existing fossil fuels, oil shale is one of the most promising energy resource with great potential in exploration and development. However, oil shale entrained large amount of inorganic impurities during the shale formation and mining process[4]. This reduces the grade and grindability of oil shale, which limits the development and utilization of oil shale. Comminution is the first step in coal and oil shale beneficiation process and has been proven to liberate organic and inorganic matters effectively [5-7]. Any pretreatment technique that can increase the grindability by inducing structural defects would lower the energy required by a mill to achieve the same size class. The importance of pretreatment is emphasized not just because of mitigation of energy intensity. A finer pulverised oil shale can be further upgraded with other beneficiation processes, like floatation [7, 8] and separation [9] to obtain a higher oil yield [10, 11].

The previous works on coal beneficiation provided insights that are relevant to oil shale pretreatment. However, the mechanical and chemical properties of oil shale determine the suitability of the pretreatment methods. The variable properties of oil shale such as moisture content, composition and grade can all affect its hardness. Unlike coal, the effects of moisture content on oil shale grindability is contradictory. Clay minerals and quartz are the dominating minerals, which increases oil shale's affinity for hydration [12]. When water or other fluid permeate into the shale, the cohesion between layers will decrease and the hydraulic wedge effect will lead to defects in structure. However, this is not an economically viable method due to the energy required for drying or dehydration after inducing water. Conventional preheating can reduce grinding resistance of coal and oil shale [13-15]. Conventional heating is also called resistive heating. Heat is transferred to the solid fuel by mechanisms of conduction, radiation and convection [16]. Consequently, the material's outer layer is heated first followed by the heat transferred towards the inner layers. Hence, the process highly depends on thermal conductivity of the material and requires time. Also, prolonged heating duration may lead to unexpected impacts on material (e.g. oxidation). These drawbacks make conventional preheating method less attractive.

Microwave heating is unique and delivers several advantages over conventional heating, such as rapid heating, material selective heating, quick start and stop response [17]. Microwave energy, as an electromagnetic energy, is absorbed by target object volumetrically at a molecule level [18] and results in uniform internal and external heating simultaneously. Still, the microwave absorbing ability is highly depended on dissipation factor, which is the ratio of loss factor (ε'') and dielectric constant (ε). For non-homogeneous ores, minerals have different dissipation factor and thermal properties. When microwave energy is applied, selective heating occurs and induces thermal stress cracking between different phases due to significantly different thermal expansion coefficients [19]. Previous work has shown oil shale, like ores, also responds to microwave and the grindability can be positively affected.

This study presents the effects of microwave-enhanced pretreatment on the nature of oil shale in comparison with conventional drying process. The milling performance of treated and untreated oil shale samples were quantitatively evaluated in terms of uniaxial compressive strength (σ_{max}) and breakage rate (S_i). Thermogravimetry and FTIR analysis was also implemented to determine any changes in chemical characteristics.

2. Materials and Methods

2.1. Material specifications

Two Chinese oil shale from Fushun (Jilin Province, China) and Xingsheng (Heilongjiang Province, China) deposits were selected for this study due to their abundant availability in China. These samples were prepared according to International Standard ISO 13909 and grounded into size fraction of 1mm-1.18mm. In addition, oil shale samples were cut into a 1.37cm×1.37cm×1.4cm cube-shaped specimens (approximate 6.4g) using a diamond coated saw blades. The specimens were drilled from the same oil shale sample and cut with the long axis perpendicular to the bedding plane of oil shale formation.

Proximate analysis was conducted via Thermogravimetric Analyser (TGA) (Netzsch STA449F3, German), while the elemental analysis was conducted with an Elemental Analyzer (Euro Vector EA3000, Italy). The oil yield of two oil shale samples were determined using a standard Fischer Assay (F.A.) oil yield test following the Chinese Standard SH/T 0508-92. Mineral composition and major mineral phase of oil shale was determined using SEM/EDS (Zeiss Sigma VP, German) and X-ray Diffraction (XRD) spectrometer (Bruker D8 Advance, German). The chemical properties and mineral composition were summarized in Table 1, while the FTIR was used to identify any changes on the surface of oil shale (Bruker Vertex 70, German).

Table 1. Chemical properties of selected oil shale samples

Properties	Fushun	Xingsheng
Proximate analysis (wt%)		
Moisture	1.34	2.20
Volatile	18.03	13.63
Fixed Carbon	2.39	17.33
Ash	78.24	66.84
Ultimate analysis (wt%)		
C	47.23	63.02
H	6.06	4.33
O	35.95	29.54
N	6.79	1.88
S	3.97	1.23
Fischer Assay (wt%)		
Water	1.7	2.3
Oil yield	10.7	5.7
Spent shale	84.3	90.1
Gas+loss	3.3	1.9
Mineral composition (wt%)		
Al_2O_3	26.5	26.7
SiO_2	56.9	63.6
FeS_2	9.7	2.4
K_2O	2.6	2.6
TiO_2	1.6	2.1
MgO	1.8	0.5
CaO	0.9	2.1

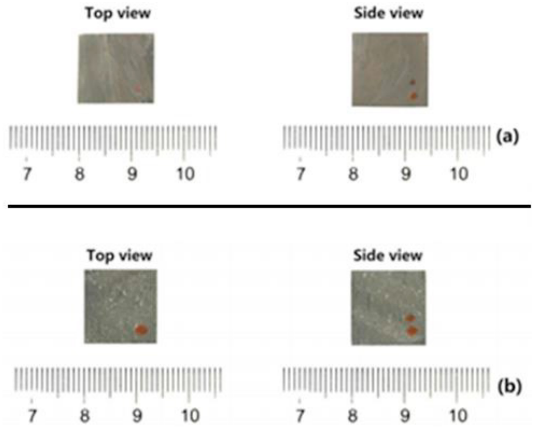

Fig.1. Optic images of Fushun (a) and Xingsheng (b) raw sample

2.2. Conventional and microwave-enhanced pretreatment methods

Microwave-enhanced pretreatment of oil shale was conducted in a 3kW and 2.45GHz multimode microwave reactor (Nanjing Jiequan Microwave Co., Ltd., China). Representative oil shale sample (1-1.18mm) and prepared specimens were exposed in microwave cavity at 1kW for 20s and 30s respectively. Conventional preheating was conducted in a horizontal tube furnace (SG-GL1200K, China). The samples were heated from 30^oC to 150^oC and 250^oC with a heating rate of 15^oC/min. After all tests, the treated sample

2.3. Uniaxial compressive strength and grindability test

A Sans 600kn universal test machine was used to determine the uniaxial compressive strength (MTS Industrial systems, Co., Ltd., China). The rate of force loading is 100N/s and the drop rate is set to 80%. A lab-scale pulverizer was used to grind two oil shale samples, both untreated and treated (Xinyuan Dy500, China). The mass retained in the required size range (0.5mm-1mm) was determined and the remnants were reused for further crushing. The weight was recorded after 0, 5, 10, 15, 20, 25, 30, and 35s.

3. Results and Discussion

3.1. Characterization of untreated and treated oil shale samples

Thermal analysis results are summarized in Table 1. The high ash content of Fushun and Xingsheng oil shale implies that mechanical and thermal properties of minerals are the key factors that affect the grindability of oil shale. Based on XRD and EDS analysis, quartz and clay minerals are the dominating inorganic phases in these two samples. Previous work showed that moisture and pyrite can be heated very quickly in microwave field compared to the rock matrix surrounds them [17, 19]. For example, pyrite can achieve 1019°C in 6.75mins, while quartz can only reach 79°C [17, 19]. The rapid phase change of water in capillary holes and thermal expansion of non-carbonaceous components could induce cracks and fissures via physical and thermal stress. Thus, microwave heating has exhibited positive effects on oil shale's grindability. However, it is vital to minimize the negative influences on the chemical properties of oil shale during pretreatment. Table 2 showed that the overall proximate composition of treated oil shale samples is comparable with untreated sample, even though some light volatiles might release via thermal degradation. The FTIR spectra in Fig. 3 are in good agreement with proximate analysis. It indicates that the organic components in Fushun and Xingsheng oil shale are similar, which showed a pronounced aromatic matrix. Additionally, the peaks at 2924cm^{-1} and 2853cm^{-1} in Fushun oil shale become more intense, which suggests the slightly change of chemical structure during the pretreatment.

Table 2. The proximate analysis of all tested samples (wt%, as received)

	Fushun oil shale				
	Raw sample	150ºC	250ºC	1kw, 20s	1kw, 30s
Moisture	1.34	0.59	0.22	1.16	1.09
Volatile matter	18.03	18.21	18.07	18.11	18.04
Fixed Carbon	2.39	2.56	2.38	2.49	2.44
Ash	78.24	78.64	79.33	78.24	78.43
Fuel Ratio	0.13	0.14	0.13	0.14	0.14
	Xingsheng oil shale				
	Raw sample	150ºC	250ºC	1kw, 20s	1kw, 30s
Moisture	2.20	1.17	0.31	1.92	1.86
Volatile matter	13.63	13.75	13.92	13.83	13.80
Fixed Carbon	17.33	17.83	17.58	17.34	17.32
Ash	66.84	67.25	68.19	66.91	67.02
Fuel Ratio	1.27	1.30	1.26	1.25	1.26

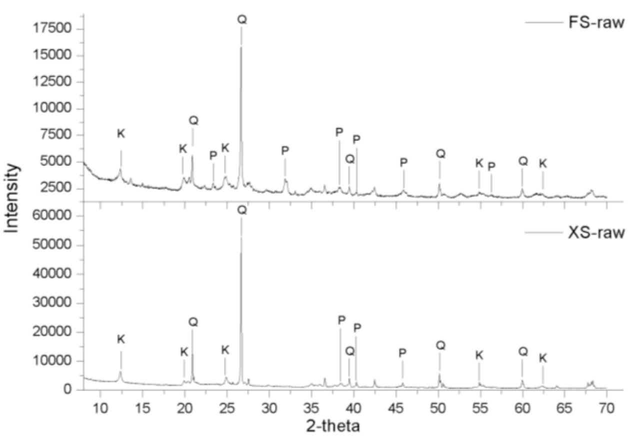

Fig. 2. Identification of major mineral phase in Fushun and Xingsheng oil shale. K-Kaolinite; Q-Quartz; P-Pyrite

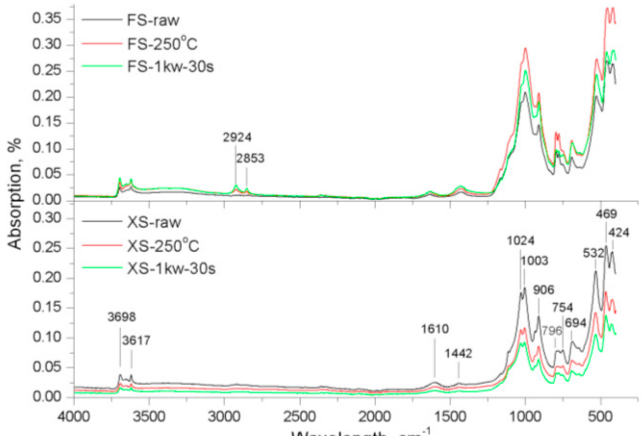

Fig.3. FTIR spectra of Fushun and Xingsheng oil shale before and after pretreatment.

3.2. Evaluation of milling performance of treated oil shale samples

The oil shale milling performance can be evaluated via the changes in maximum compressive strength (σ_{max}) and breakage rate (S_i). For conventional preheating at 250°C, maximum strength (σ_{max}) decrease 41.2% and 37.1% for Fushun and Xingsheng oil shale respectively. However, microwave-heating led to a total 63.1% reduction in maximum strength (σ_{max}) of Fushun oil shale compared with 34% decrement of Xingsheng oil shale. It is due to that microwave heating has high selectivity and the pyrite distributed in microwave-transparent rock matrix is heated and expanded rapidly, which can induce significant thermal stress. To some extent, the pyrite content has a positive effect on oil shale grindability during microwave-enhanced pretreatment.

Breakage rate can be modelled using first order kinetics when only consideration one size friction [15]. The expression of the model is shown in Eq. (1). Thus, the breakage rate (S_i) can be determined by plotting natural logarithm of the mass of samples above size class i against grinding time. Table 2 showed that the model is well-fit for the experimental data. The increase in breakage rate indicates that less energy is required by the pulverizer to achieve the same size class. It is noticeable that the specific breakage rate of Fushun oil shale increased 44.9% via exposure in 1kw microwave field for only 30 seconds, which is more competitive than conventional heating with respect to energy consumption and potential changes in chemical properties, such as loss of volatiles and thermal degradation.

$$\ln \frac{X_i(t)}{X_i(0)} = s_i t \quad (1)$$

Where $X_i(0)$ is the amount of size i material at time 0;
$X_i(t)$ is the amount of size i material at time t;
S_i is the specific breakage rate;
t refers to milling time;

Table 3. The changes in Peak strength and breakage rate of all tested samples

Specimens	Peak strength (MPa)	Reduction in strength	Breakage rate (S_i)	Increase in Grindability	R^2
FS-raw	84.78	-	-0.0127	-	0.9984
FS-150ºC	72.84	14.1%	-0.0138	8.7%	0.9824
FS-250ºC	49.85	41.2%	-0.0163	28.3%	0.9951
FS-1kw-20s	37.11	56.2%	-0.0176	38.6%	0.9947
FS-1kw-30s	31.32	63.1%	-0.0184	44.9%	0.9869
XS-raw	46.86	-	-0.0396	-	0.9984
XS-150	37.70	19.5%	-0.0427	7.8%	0.9768
XS-250ºC	29.48	37.1%	-0.0438	10.6%	0.9984
XS-1kw-20s	38.27	18. 3%	-0.0427	7.8%	0.9869
XS-1kw-30s	30.95	34.0%	-0.0436	10.1%	0.9989

Conclusion and future work

This paper investigates the effects of microwave-enhanced pretreatment on the nature of oil shale compared with conventional drying process. The milling performance of treated oil shale samples was significantly improved in terms of decrease in peak uniaxial compressive strength (σ_{max}) and increase in breakage rate (S_i). The proximate analysis and FTIR spectra indicate that the proposed pretreatment methods have negligible impacts on chemical properties of oil shale particles.

The high efficiency of microwave treatment on oil shale milling performance is illustrated by the different dielectric properties and thermal expansion coefficients of different mineral phases in oil shale. In this case, the reduction of peak strength of Fushun oil shale is twice larger than that of Xingsheng oil shale under identical pretreatment due to the different mineral compositions. Therefore, it is more plausible that the cracks in inorganic phases are responsible for the promoting effects of microwave pretreatment

The impacts of microwave pretreatment on moisture content are negligible within a short exposure time. However, the rapid phase changes and expansion of water in small pores are likely to yield physical stress on the pore structure, while moisture itself may experience several times phase changes before releasing from a dense rock matrix. If this is the case, moisture is another factor that affects oil shale grindability during microwave-enhanced pretreatment. Further experiments are required to draw the conclusion.

Additionally, it is noticeable that breakage rates increase with the reduction of peak strengths. However, the changes in peak strength are more significant. Future work will be focused on the impacts of particles size on the performance of microwave on oil shale samples, which is vital for the application of microwave pretreatment approach and optimization of microwave cavity during industrialization.

Acknowledgements

This work was supported by the Natural Science Foundation China (Grant No.51650110508).

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