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Physicochemical properties of Indonesian lignite continuously modified in a tunnel-type microwave oven for slurribility improvement

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ABSTRACT

Indonesian lignite was continuously modified in a tunnel-type microwave oven to improve its slurribility. The maximum solid concentration of coal–water slurry (CWS) prepared from modified lignite increased from 41.6 wt.% to 54.0 wt.% with an enhanced rheological behaviour. The contact angle, oxygen functional groups, molar ratio of methylene to methyl, paramagnetic centres of modified lignite were analysed on contact angle analyzer, Fourier transform-infrared spectroscopy, X-ray photoelectron spectroscopy, electron paramagnetic resonance spectroscopy to clarify the modification mechanisms. The molar ratio of the methyl group to the methylene group of the modified lignite decreased, resulting in shortened aliphatic chains and condensed aromatic rings. The unpaired electrons concentration of the modified lignite increased, implying that the coal rank was upgraded. As a result, the contact angle of the modified lignite increased and oxygen functional groups with high hydrophilicity decreased, leading to an improved CWS property.

1. Introduction

Coal–water slurry (CWS) as a promising substitute fuel for petroleum has been widely used in boilers and gasifiers [1–5]. CWS fuel has been used in over 20 power plants, over 300 industrial furnaces and several hundreds of various kilns in China. The largest full CWS-fired 670 t/h boiler (200 MW) has been successfully operated in Nanhai power station in Guangdong Province of China since 2005 [6]. Over 35 large-scale CWS gasifiers (CWS consumption amount per gasifier: 750–3000 t/d) have been commercially operated in China. The largest CWS gasifier with CWS consumption of 3000 t/d has been put into operation in Rongxin Chemical Company in Inner Mongolia since June 2014. It is estimated that about 30 million tons of CWS fuel were consumed in boilers and gasifiers in China every year. CWS is composed of 50 wt.% to 75 wt.% pulverised coal, 25 wt.% to 50 wt.% water and <1 wt.% chemical additives [7]. The characteristics of CWS are influenced by the coalification degree, chemical compositions (viz. inherent moisture, oxygen functional groups), wettability, particle size distribution, pore structure distribution and chemical additives.

Lignite occupies 45% of the world's coal reserves [8]. Lignite has been widely used in many boilers in mine-mouth power stations, because lignite with high moisture and low heating value is not suitable for long-distance transportation. Lignite with high reactivity is deemed ideal for gasification and has been used in many gasifiers [9]. CWS prepared from lignite provide alternate application routes in boilers and gasifiers, because it can be conveniently and cleanly stored, transported, atomised and burned like heavy oil. Producing high-quality CWS fuel with a high concentration (55 wt.% to 70 wt.%) and low viscosity (<1200 mPa·s at a shear rate of 100 s^{-1}) from lignite used for combustion and gasification is challenging because of the high inherent moisture content and oxygen content, poor grindability and low coalification degree of lignite. The maximum solid concentration of CWS prepared from lignite is low (40–45 wt.%) [10]. Thermal heating (100–900 °C, 0.1 MPa) [11–13], fluidized-bed drying process (120–300 °C, 0.1 MPa) [14–17], hydrothermal treatment (250–350 °C, saturated vapour pressure) [18–23], mechanical thermal expression (180–200 °C, 4–6 MPa) [23–25] and upgraded brown coal (UBC) process based on slurry dewatering technology (130–160 °C, 0.35–0.45 MPa) [26–28] have been employed to upgrade lignite. However, the industrial applications of these techniques are limited because of their high energy consumption and complicated operation.

Microwave irradiation has some advantages over conventional heating methods [29]. These advantages include rapid heating, quick turn on or off, selective heating, uniform and volumetric heating and energy transfer instead of heat transfer. Thus, microwave irradiation is an effective modification process for coal. A number of potential applications of microwave irradiation on coal

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Nomenclature

M_{ad}	moisture on an air dry basis	CH_2/CH_3	methylene to methyl ratio
A_{ad}	ash on an air dry basis	O/C	atom ratio of oxygen to carbon
V_{daf}	volatile on a dry and ash-free basis	C—C/C—H	graphitized carbon
FC_{daf}	fixed carbon on a dry and ash-free basis	C—O	carbon in phenolic or alcohol group
$Q_{net,ad}$	net calorific value on an air dry basis	O=C—O	carbon in carboxyl or ester group
CWS	coal water slurry	C=O/O–C—O	carbon in carbonyl or quinine group
XPS	X-ray photoelectron spectroscopy	NDF	copolymers of sodium methylene naphthalene sulphonate– sodium styrene sulphonate–sodium maleate
FTIR	Fourier transform-infrared spectroscopy		
EPR	electron paramagnetic resonance spectroscopy		
HI	hydrophilicity index		

treatment have been investigated and verified, including dewatering [8,30], upgrading [29], desulphurisation [31], increased grindability [32–35], improved floatability [36], rapid coke making [37], pyrolysis [38] and improved oil yield through liquefaction [39] during coal treatment. The unit energy consumptions for CWS concentration promotion and inherent moisture removal by thermal heat are respectively 214 and 22.5 times higher than by microwave irradiation [11]. Microwave irradiation is more promising and cost-efficient in CWS property improvement than thermal heating.

To abate erosion during pipeline transport, Meikap [40] investigated the selective demineralisation effect of microwave irradiation (microwave power of 550 W, irradiation time of 60 s, sample mass of 500 g, N_2 atmosphere) to reduce the slurry viscosity and increase the solid concentration of high-ash Indian coal. This investigation revealed that the solid concentration is low at 20–40 wt.%, the viscosity is lower than 300 mPa·s at a shear rate of 20 s^{-1}, and the particle size is 60–250 μm. Sakoo [41,42] found that microwave-treated high ash Indian coal slurry (microwave power of 900 W, four irradiation time levels of 30–120 s, sample mass of 500 g, N_2 atmosphere) enhanced rheological properties and abates erosion during pipeline transport. This was probably because microwave treatment resulted in pyrite decomposition and α-silica conversion into β-silica. This study also determined that the solid concentration is low at 30–50 wt.%, the viscosity is lower than 200 mPa·s at a shear rate of 20 s^{-1}, and the particle size is 60–253 μm. Cheng [11] improved the properties of CWS prepared from air-dried pulverized Shenhua coal by employing microwave irradiation (microwave power of 280–700 W, irradiation time of 20–60 s, sample mass of 8 g, N_2 atmosphere) and further analysed the chemical compositions (proximate and ultimate analyses) and pore structures of the modified coal. They suggested that microwave heat was much better than thermal heat in coal upgrading to improve CWS property. However, the microwave modification of coal in these studies was conducted on small- and batch-scale microwave units under a power lower than 900 W, an irradiation time shorter than 120 s and a coal mass less than 500 g. These conditions are insufficient to meet industrial- and commercial-scale requirements for CWS volume production.

However, the effects of microwave irradiation on specific physicochemical properties of lignite (viz. surface wettability, oxygen functional groups, aliphatic chains, free radicals, and chemical additive adsorption) have been seldom investigated in the literature. Furthermore, the effects of microwave irradiation on lignite upgrading for slurribility improvement on a continuously tunnel-type microwave oven remain unclear. Therefore, the present study modified Indonesian lignite on a continuous tunnel-type microwave irradiation system for slurribility improvement. The microstructures of the modified lignite were investigated by contact angle analysis, Fourier transform-infrared spectroscopy (FTIR), X-ray photoelectron spectroscopy (XPS), electron paramagnetic resonance spectroscopy (EPR), laser particle size analysis and rotary viscometry to prepare high-quality CWS.

2. Material and methods

2.1. Material

Indonesian lignite was modified by microwave irradiation. The proximate analysis was measured in accordance with ISO 11722, ISO 1171 and ISO 562. The elemental analysis was measured in accordance with ISO 625, ISO 333 and ISO 334. Calorific value analysis was conducted with the bomb calorimetric method in accordance with ISO 1928. High-quality CWS is difficult to derive from Indonesian lignite because of its high inherent moisture.

2.2. Experimental installation

Indonesian lignite was continuously modified in a tunnel-type microwave irradiation system. The tunnel-type microwave irradiation system (5800 mm × 1200 mm × 1600 mm) was principally composed of two multimode resonant cavities. Each cavity was equipped with three sets of 1 kW microwave generators (single magnetron power, 1 kW; frequency, 2450 ± 50 MHz) and an infrared temperature measuring point (monitor temperature to prevent spontaneous combustion in lignite seam). The microwave generated by each magnetron was transmitted through the waveguide tube and fed into the resonant cavity from its top. Electromagnetic suppressors with a height of 40 mm were placed on both sides of the cavities to prevent microwave leakage. Lignite (particle size less than 25 mm) was continuously transported by a polytetrafluoroethene conveyor belt (width, 400 mm) passed through electromagnetic suppressors and resonant cavities. Produced steam and other exhaust gases were emitted by fans. The microwave power was regulated at 3–6 kW. Lignite (spreading width, 320 mm; thickness, 25 mm) was continuously transported at a certain speed with a constant effective microwave irradiation time of 40 min. The modified lignite was pulverised to less than 150 μm. CWS was prepared by mixing pulverised coal, deionised water and NDF chemical additive [18] (1 wt.% based on air-dried pulverised coal weight).

2.3. Methods

2.3.1. Contact angle of modified coal

The contact angle was geometrically defined as the angle formed by a liquid at the three-phase boundary where a liquid, gas, and solid intersect. 1 g pulverized lignite was pressed into a cylinder with 20 mm diameter and 2 mm thickness at 10 MPa for 30 min. A pendant droplet of distilled water was deposited on the cylinder lignite sample. The contact angle of modified lignite