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Bioliquefaction of the extracts from Shengli lignite

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ABSTRACT

Shengli lignite was sequentially extracted with aqueous NaOH solution, *n*-hexane, dichloromethane (DCM), isometric benzene and ethanol (IMBE), and acetone to obtain extracts 1–5 (E_1–E_5). Each extract was biotreated by an isolated fungus WF8 under the same conditions. The results show that E_1 is much more susceptible to the bioliquefaction (BL) than other extracts. The BL of E_1 proceeded via the cleavage of C–O–C and Si–O bonds to some extent, while most of the methylaromatic and –COOH-containing moieties in E_1 are inert toward BL by WF8. E_1 has largely similar molecular mass distribution (MMD) to unliquefied E_1, but quite different MMD from bioliquefied E_1, further indicating that the BL of E_1 proceeded via biochemical reactions by WF8.

1. Introduction

Thermochemical conversion processes of coals under severe conditions usually require huge investment and even pose a serious potential threat to the environment [1–7]. This is especially true for lignites with low calorific value and high ash content. In contrary, bio-treatment of coals by microorganisms could proceed under mild conditions. Microbial treatment of lignites has been considered as a cost-effective and ecofriendly way for converting coals to clean fuels and value-added chemicals [8,9]. A number of microorganisms were tested for coal bioconversion to obtain liquid or gaseous products [10–18].

However, very complex organic matter in lignites is rather resistive to direct bioliquefaction by microorganisms, resulting in low conversion and low microbial efficiency. Oxidative pretreatment can improve bioliquefaction of lignites, but such a pretreatment leads to the introduction of some undesired groups [19,20]. As a nondestructive and separable method, fractional extraction was widely used to isolate trace species [21,22] and group components [23,24] in organic matter from coals and their derivatives. Compared to coal matrix, coal extracts have much simpler compositions and thereby can be analyzed more easily.

In the present work, we isolated the complex organic matter in Shengli lignite (SL) to several extracts by fractional extraction and examined the bioliquefaction (BL) of the extracts.

2. Experimental

2.1. SL, solvents, and fungus

SL was freshly collected from Shengli Coal Mine, Inner Mongolian Autonomous Region of China. It was pulverized to pass through a 90-mesh sieve (< 0.16 mm) followed by desiccation in an oven at 80 °C for 24 h before use. All the organic solvents used are analytical reagents and were distilled before use with a rotary evaporator. The fungus used was isolated from decaying wood around coal mines and designated as WF8, which can excrete some ligninolytic enzymes (Fig. S1). It was tentatively identified as *Ascomycota Hypocera lixii* on the basis of its 18S rDNA gene sequence (GenBank accession No. JQ806366). It is stored on Subaroud agar slants at 4 °C.

2.2. Fractional extraction

About 10 g SL was steeped into 150 mL 0.2 mol·L^{-1} NaOH solution at 60 °C for 24 h, then centrifuged at 8000 rpm for 15 min. The supernatant was precipitated by adjusting pH to 2.0 with 6 mol·L^{-1} HCl to obtain extract 1 (E_1), which was dried in an oven at 80 °C, weighed, and stored in a vacuum-dried chamber.

As shown in Fig. 1, the residue was sequentially extracted with 150 mL of *n*-hexane, dichloromethane (DCM), isometric benzene and ethanol (IMBE), and acetone in a modified Soxhlet extractor with a water bath. Each extraction was conducted at least 4 days [21,25] to extract the soluble portion as exhaustively as possible. The

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Nomenclature

IMBE	isometric benzene and ethanol
BL	bioliquefaction
BLE_1–BLE_5	bioliquefied extracts 1–5
CAs	carboxylic acids
DART	direct analysis in real time
DART/MS	direct analysis in real time/mass spectrometer
DCM	dichloromethane
E_1–E_5	extracts 1–5
FTIR	Fourier transform infrared
GC/MS	gas chromatograph/mass spectrometer
MMs	molecular masses
MMD	molecular mass distribution
NAs	normal alkanes
OCs	organic compounds
ONs	organonitrogens
PCs	phenolic compounds
RAs	relative abundances
SL	Shengli lignite
ULE_1–ULE_5	unliquefied E_1–E_5

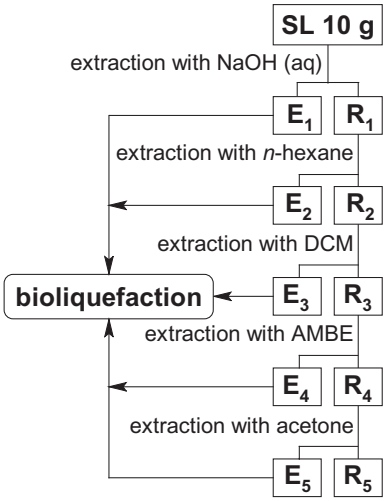

Fig. 1. Procedure for the fractional extraction of SL and subsequent bioliquefaction of the extracts.

temperatures of the solvents dropping onto SL in extraction thimble from a condenser are lower than 60 °C. Each extract was concentrated by removing the solvent using a rotary evaporator to obtain extracts 2–5 (E_2–E_5).

2.3. BL of the extracts

WF8 was inoculated onto the Subaroud plate medium (40 g maltose, 10 g peptone, 20 g agar, and 1 L distilled water) in Petri dishes of 6 cm diameter, followed by incubation in a biological incubator at 28 °C for ca. 3 days. After the development of mycelia on the medium surface, 0.2 g of each extract (E_1–E_5) was manually well distributed onto the dishes, respectively. After 7 days, black liquid droplets were formed on the surface. The liquid products, i.e., bioliquefied E_1–E_5 (BLE_1–BLE_5), were collected using a finnpipette and stored in a sample bottle at 4 °C for subsequent analysis. The residues of extracts, i.e., unliquefied E_1–E_5 (ULE_1–ULE_5), were washed and the resulting suspension liquids were centrifuged at 8000 rpm for 10 min to remove the WF8 mycelia as exhaustively as possible followed by drying the precipitates in an oven at 80 °C for 12 h and storing the dried precipitates in a desiccator at 4 °C. Media with WF8 alone and with each extract alone were as controls.

2.4. Sample analysis

The analyses of selected samples were conducted with a Nicolet Avatar 380 Fourier transform infrared (FTIR) spectrometer using KBr pellets with a resolution of 4 cm^{-1} in the range of 4000–400 cm^{-1}, Leco Mac-400 thermogravimetric analyzer, Leco CHN-2000 elemental determinator, Leco SC-132 sulfur determinator, Agilent 7890/5975 gas chromatograph/mass spectrometer (GC/MS) equipped with a capillary column coated with HP-5MS (60 m × 0.25 mm × 0.25 μm) and a quadrupole analyzer and operated in electron impact (70 eV) mode, and IonSense direct analysis in real time/mass spectrometer interfaced to a mass spectrometer (DART/MS) using He as the discharge gas and nitrogen N_2 as an alternative gas with a flow rate of 2 $L{\cdot}min^{-1}$ and operated at 450 °C.

3. Results and discussion

3.1. BL of E_1–E_5

E_1 is recognized as humic acids and E_2–E_5 consist of normal alkanes (NAs), arenes, alkanols, phenolic compounds (PCs), ketones, carboxylic acids (CAs), esters, and organonitrogens (ONs) according to analysis with GC/MS shown in Figs. S2–S5 and Tables S1–S8 [7]. Among the extracts, E_5 is extremely viscous and cannot be uniformly distributed on the plate medium. Therefore, BL cannot be performed on E_5. As Fig. 2

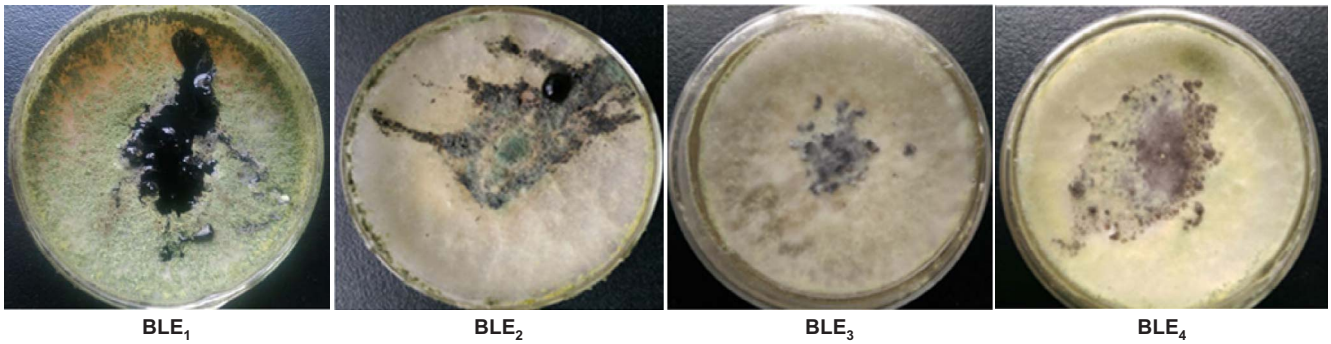

Fig. 2. Photographs of BLE_1–BLE_4 by WF8 after 7 days.