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Research article

Dielectric characterization of Indonesian low-rank coal for microwave processing

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ABSTRACT

The present study aims to provide a preliminary guide for microwave processing of low-rank coals by investigating the dielectric characteristics of an Indonesian low-rank coal based on determination of its permittivity from 21 °C to approximately 1000 °C at 915 and 2450 MHz in argon. The results show that the relative dielectric constant of the coal gradually decreases with increasing temperature below 200 °C due to dewatering. It then has a considerable increase up to 750 °C because of devolatilization and carbonization, preceded by a slight decrease due to physical-chemical and chemical processes connected with the mobility of macromolecular chains of the coal. Thereafter, it increases with a declined rate ascribed to the coal's continuous structure transformations. The dielectric loss factor shows a similar variation, except the presence of the "dielectric loss peak" at about 600 °C, associated with rise of microwave loss. The microwave penetration depth of the coal presents a depth peak at approximately 400 °C, indicating potential application of variable frequency microwave heating to large-scale processing. The reflection loss (*RL*) patterns reveal the strong coal slab thickness dependences of magnitude and location of absorption peaks in the range 0.02–0.20 m, with minimum *RL* values of −42.39 dB and −47.92 dB obtained at thicknesses of 0.16 and 0.06 m at 915 and 2450 MHz, respectively. This dissimilarity is primarily attributed to the different dielectric parameters and wavelengths at these frequencies. Microwave heating functions sufficiently in a narrow temperature range from about 500 to 700 °C, in which target temperature can be selected for efficient coal processing.

1. Introduction

Coal is one of the most important fossil fuels constituting 30.0% of global primary energy consumption [1]. With continuous consumption and gradual depletion of high-rank coals, the demand for utilization of low-rank coals, such as lignite which accounts for almost half of the world's coal reserve [2], becomes increasingly stringent. It is promising to exploit these low-rank coals because they offer a wide range of competitive advantages, including low mining cost, good reactivity, high volatile matter and low pollutant impurities (e.g., sulfur, nitrogen, and heavy metals) contents [3]. However, due to their higher transportation/storage costs, lower efficiency, greater CO_2 emissions and higher spontaneous combustion tendency compared to high-rank coals, use of low-rank coals still faces great technical challenges [4].

Regarding these above problems, many novel techniques were developed to upgrade low-rank coals or to convert them to more valuable products. As a new volumetric heating technique, microwave radiation has been widely used for the treatment of coal [5,6]. It is characterized by rapid and selective heating, uniform heat distribution, flexible modular design, easy and instant operation (switch-on and switch-off), and environmental friendliness [7]. With these unique characteristics, microwaves enable a large number of relevant applications, including coal drying/dewatering [8], grinding [9], floatation [10], coal-water slurry combustion [11], desulfurization [12,13], coking [14], pyrolysis [15] and liquefaction [16].

For low-rank coals, microwave drying and pyrolysis have received exceptionally intensive attentions in the past decade [17]. Specifically, owing to the internal and selective heating features of microwaves, the rate of moisture removal of microwave-assisted coal drying was found to be one to two orders of magnitude faster than that of conventional convective drying [18]. The rapid moisture removal also contributed to a substantially lower unit energy consumption (decreased by approximately 22.5 times) of the process compared to conventional thermal heating [19]. It was also revealed that the microwave drying process led to higher calorific value, greater fixed carbon content,

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increased pore volume, enlarged surface area, increased ignition and burnout temperatures, and decreased comprehensive combustion parameter of low-rank coals [20]. From the results of the oxygen/carbon ratio parameter verified by the Fourier transform infrared spectroscopy, low-rank coals were upgraded to high-rank coals after the drying process. The changes in physicochemical characteristics became more notable with increasing processing temperature (e.g., from 130 °C to 160 °C). Microwave treatment was thus considered to be an effective dewatering process [20]. As another important application, microwave pyrolysis exhibits many superior features. The internal heating of microwave enhances the heating rate of coal particle during pyrolysis, leading to a faster escaping rate of volatile matter out of coal particles, which eventually leads to a decrease in the secondary reactions of volatile matter and to an increase in oil yield [15]. The process can be further improved through addition of susceptors, including graphite [21], coke [22] and microwave-absorbing metal oxides (e.g., Fe_3O_4 [15], CuO, V_2O_5 and WO_3 [22]). This operation not only significantly reduced the processing time of low-rank coals (e.g., Indonesian coal), but also improved the quality of pyrolysis products, such as coal oils with increased high heating value (HHV, 33–39 MJ kg^{-1}) [21]. After mild upgradation, these products could potentially replace conventional fuels. Overall, it is feasible to control the pyrolysis process precisely to obtain products with more desirable properties by taking advantage of uniform temperature distribution, decreased secondary reactions and internal heating of feedstock involved in microwave pyrolysis [23].

Although use of microwave has shown great potential in coal processing, many of above-mentioned processes still confront strict energy requirements, especially for industrial/large-scale applications in a broad temperature range. This is because in large-scale uses at elevated temperatures microwaves in reaction cavity may fail to penetrate sufficiently far into the coal which may cause inhomogeneous heating or loss of control associated with serious thermal runaway [24,25]. To prevent this problem for smooth operation and high processing efficiency, appropriate design of reaction cavity has been deemed a crucial solution [26]. The optimal cavity should have dimensions compatible with microwave amplitude diminishing (and thus wave dissipation) as a consequence of power absorption as heat within the sample [27]. From this perspective, microwave dissipation behavior in coal should be determined first. Such behavior, without doubt, depends strongly on the coal's own properties, namely dielectric properties which vary with a range of factors, including coal rank, moisture content, temperature and applied frequency [5,28].

Considering the importance of dielectric properties of coal, several relevant characterization studies have been carried out. In general, coal as a bulk material at low temperature has relatively poor dielectric responses while coal constituents such as moisture [29,30], bound hydroxyl groups (—OH) [5] and pyrite (FeS_2) [28] have considerably higher dielectric loss than the organic components, with the organic constituent behaving as a matrix that is essentially transparent to microwaves. For example, the dielectric loss factors of a high-volatile coal (moisture 6.1%, volatile matter 34.8%, fixed carbon 55.7%, and ash 3.4%) and its organic matrix were found to be 0.1389 and 0.058 at 2 GHz and 60 °C, respectively [28], which were much lower than those of water (8.4 [30]) and pyrite (1.0625 [28]). Most of these studies showed an emphasis on the low temperature dielectric properties of coal. However, few reports explored microwave dielectric properties of coal (particularly, low-rank coals) at elevated temperatures [31]. A previous study explored the dielectric properties of a bituminous coal (moisture 1.03%, volatile matter 30.25%, fixed carbon 59.86%, and ash 9.89%) during pyrolysis from room temperature to approximately 900 °C. It was shown that the dielectric response remained relatively constant below 500 °C and then increased sharply at higher temperatures due to devolatilization and carbonization [31]. The significance of conductive loss in the microwave absorption capability of the coal was identified in this work but the changes in the microwave loss mechanism during the process (e.g., from dipolar polarization to conduction) were not illustrated. A recent work examined the variation of dielectric parameters of low-volatile anthracite in a similar temperature range. It was found that the properties increased linearly with increasing temperature, which was primarily attributed to electronic conduction and potential interfacial polarization (a type of relaxation polarization) [32].

Currently, the dielectric behavior of low-rank coals like lignite is not well understood in the literature. The present study investigates the dielectric properties of an Indonesian low-rank coal (lignite) by measuring its dielectric parameters in argon up to 1000 °C at industrial frequencies, 915 and 2450 MHz. The coal's microwave dielectric characteristics were explored by evaluating its dielectric loss tangent, microwave penetration depth and reflection loss (*RL*). The findings are expected to provide a useful preliminary guide for optimizing operating conditions (e.g., temperature and applied frequencies) of upgrading low-rank coals using microwave energy to obtain maximal products with high efficiency.

2. Experimental

2.1. Coal sample preparation and dielectric measurements

An Indonesian low-rank coal (lignite, Sumatra Lahat, Indonesia) was used for the measurement of dielectric properties. Its proximate and ultimate analyses are summarized in Table 1. For dielectric characterization, the coal sample was ground to particles having size less than 75 μm and then dried (105 °C, 15 h) in a resistance oven. These particles were then unidirectionally pressed in a tungsten carbide-coated die to prepare 3 pellets (ø 3.70 mm, a total, piled length of 13.90 mm) for successful measurement. The pressing operation aims at maintaining homogeneous microwave distribution within the sample which ensures the accuracy of dielectric properties measurement [33]. The initial pellet density was 1.04 g cm^{-3}, and the final pellet density after the experiment was 0.99 g cm^{-3}, showing weak influence of pellet density change on the measurement. The mass loss during the experiment was 52% of the initial mass, mainly due to devolatilization of the sample.

For measurements of dielectric properties of the coal, the cavity perturbation technique (CPT) was used in the present study. In CPT, the differences in the microwave cavity response, namely the frequency shift and change of quality factor, caused by insertion of sample were employed to compute the complex relative permittivity [33]. The measurement system was mainly constituted by a resistance furnace and a cylindrical TM_{0n0} resonant mode cavity with a diameter of 580 mm and a height of 50 mm. The furnace was used to heat the sample to the preset temperature and the cavity was employed to detect the abovementioned cavity response differences caused by sample at each examined frequency (915 MHz and 2450 MHz), which were subsequently recorded in a Hewlett Packard 8753B vector network analyzer (VNA). By obtaining the differences in the microwave cavity response, the complex electric susceptibility of the sample was computed and used for calculation of corresponding dielectric parameters. More details of CPT and its major apparatus are available in the literature [33–37].

In the measurements, the prepared coal pellet was heated from room temperature (21 °C) to about 1000 °C in 50 °C steps with a ramp rate of 5 °C min^{-1} using the furnace in 0.005 L min^{-1} flowing ultra-high purity argon. The dielectric parameters at each temperature and frequency were then computed. The entire process was pre-programmed and controlled by the LabVIEW software.

The dielectric parameters determined and computed in this study are: 1) relative dielectric constant and dielectric loss factor, 2) dielectric loss tangent, 3) penetration depth, and 4) reflection loss which explores the coal slab's microwave dielectric characteristics.