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Factors affecting the microwave coking of coals and the implications on microwave cavity design

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ABSTRACT

The work carried out in this paper assessed how processing conditions and feedstock affect the quality of the coke produced during microwave coke making. The aim was to gather information that would support the development of an optimised microwave coke making oven. Experiments were carried out in a non-optimised 2450 MHz cylindrical cavity. The effect of treatment time (15–120 min), power input (750 W–4.5 kW) and overall power input (1700–27,200 kWh/t) on a range of coals (semi-bituminous–anthracite) was investigated. Intrinsic reactivity, random reflectance, strength index and dielectric properties of the produced cokes were compared with those of two commercial cokes to assess the degree of coking produced in the microwave system. Overall energy input and coal rank were found to be the major factors determining the degree of coking following microwave treatment. The dependency on coal rank was attributed to the larger amount of volatiles that had to be removed from the lower ranked coals, and the increasing dielectric loss of the organic component of the coal with rank due to increased structural ordering. Longer treatment times at lower powers or shorter treatment times at higher powers are expected to produce the same degree of coking.

It was concluded that microwave coke making represents a potential step-change in the coking industry by reducing treatment times by an order of magnitude, introducing flexibility and potentially decreasing the sensitivity to quality requirement in the feedstock. The main challenges to development are the energy requirements (which will need to be significantly reduced in an optimised process) and penetration depth (which will require an innovative reactor design to maximise the advantage of using microwaves). Understanding and quantifying the rapidly changing dielectric properties of the coal and coke materials is vital in addressing both of these challenges.

1. Introduction

Over the last 100 years or so, knowledge about coke making and the slot oven design has been gradually optimised. However the fundamental process surrounding the transformation of coal to coke remains essentially the same, with the use of conventional ovens heating coal feed stocks for at least 16 h [1]. Whilst many aspects of production have been improved with changes in furnace design, long processing times have remained, mainly as a result of the poor thermal properties of the coal matrix [2]. Another limitation of the existing coke making process is the need to blend coals to within a fairly narrow volatile content range in order to control pressure in the oven: some pressure between the coal particles is desirable, as it improves coke strength, but excessive pressures cause damage to the oven walls [3].

World steel production has increased dramatically over the past few years. Between 1970 and 2006 production almost doubled from less than 600 Mt to around 1.2 billion tonnes [4]. Almost 70% of steel produced relies on metallurgical coal (coking coal) [4]. In addition, current coke making facilities are in need of refurbishment i.e. in 2004, 52% of factories were over 20 years old and 26% were over 30 years old [5]. The increasing demand for coke, coupled with ageing coking facilities, may open investment opportunities to new coke making methods.

Microwave heating of coal has been identified as having the potential to offer a step change improvement in coke making. Microwaves heat volumetrically, meaning that non-metallic materials, like coal, can be heated effectively instantaneously, avoiding the heat transfer limitations of conventional heating and, therefore, drastically reducing treatment times. Microwaves heat selectively, the degree of heating of each individual component depending upon its dielectric properties. Although coal as a bulk material at room temperature has relatively low dielectric properties, coal constituents such as moisture, bound hydroxyl groups and pyrite have significantly higher dielectric loss than the organic component [6–8]. There are also small but detectable

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variations in electromagnetic properties of macerals [8–10]. Bound water in porous particles has the potential to superheat to temperatures well above 100 °C [6,11–14]. This superheated water is situated within the microwave transparent coal matrix, heating it up and thereby raising the bulk temperature, increasing the susceptibility of the coal itself to microwave absorption. The transformation from coal to coke produces an increase in the aromaticity of the carbon material [7] as a consequence of the loss of the volatile component during the carbonisation and an increase in the degree of graphitisation. Increasing aromaticity results in an increase in conductivity due to increasing electron mobility through the transfer of π bond electrons along the aromatic layers [2,8–10,15–19]. As temperatures continue to increase from 400 to 1000 °C, electron mobility and level of free charge per unit volume will also continue to increase through graphitisation of the carbon, resulting in an increase in dielectric loss factor and thus increasing microwave heating, allowing the high temperatures (>1000 °C) required for coking of coal to be achieved [1,3,6,8].

Coke making by the microwave heating of coal is not a new idea, and several patents for methods with and without the use of microwave receptors have been filed, dating from the 1970s to more recently [20–23]. Coetzer and Rossouw have used microwave treatment to produce high quality cokes from a Waterberg semi-soft coking coal in 2–3 h using microwave receptors to heat the coal indirectly [24]. However, according to the theory presented above, some or even all coals could be heated to coking temperatures without the addition of microwave receptors. Since superheating of bound water is a major mechanism in the initial heating-up of the coal, it is vital to maximise the power density in the sample during cavity design. Lester et al. [6] have shown that it is possible to produce coke with similar properties to conventional cokes using a relatively low rank, high volatile bituminous coal without the use of receptors with only 70 min of heating in a multimode cavity. The cavity used was a non-optimised multimode cavity operating at 8 kW.

Initial indications from previous work are therefore that microwave heating may represent a step change in coke making through drastic reductions in coking times, and the ability to start-up and shut down the process within a short time frame. In other words, a complete redesign of coking plant equipment with a smaller footprint and the ability for flexible operation may be possible.

Prior to the development of any larger scale process, a more fundamental understanding of the microwave induced coking process is required. The importance of high temperature properties of coke is well established in the steel industry [25]. Coking conditions (such as bulk density, coking time, preheating of the charge, and the incorporation of non-coal materials) and the properties of the input coal or coal blend are the factors that determine the coke properties in conventional coking [25]. The aim of the work presented in this paper is therefore to investigate the effect of coking conditions and input coal properties on the quality of coke produced using microwave heating. The basic process variables in a microwave process are the power input and treatment time. A certain amount of input power is absorbed by the sample, and if this is measured over the duration of the experiments, the energy input can be calculated. As stated above, the input coal or blend is also a major determinant of coke quality. The coal rank, rheology and composition all affect the final coke quality [25]. In microwave processes, the dielectric properties of the material are also key factors in determining how a material will heat, and these change with temperature, pressure and composition [26]. This feeds back into the degree and distribution of power absorption, which can be expressed by the power density in the sample, as shown in Eq. (1).

$$P_d = 2\pi f \varepsilon_0 \varepsilon'' |E|^2. \quad (1)$$

The power density, P_d, is the power dissipated within a given volume of material (W/m^3), f is the frequency of the applied electromagnetic wave (Hz), ε_o is the permittivity of free space (8.85×10^{-12} F/m), ε'' is the dielectric loss factor and E is the electric field strength (V/m). P_d varies spatially and according to the cavity and sample dimensions and absorbed power, and these variations are accounted for in the E term. It is therefore clear that there is a complex interdependency of process variables and material properties, and that the heat transfer properties that determine heating rate in conventional coking are largely replaced with the dielectric properties in microwave heating.

The dielectric properties also impact reactor sizing. As a wave progresses into a dielectric-heating workload, its amplitude diminishes owing to absorption of power as heat into the material. This attenuation is expressed quantitatively by the penetration depth, D_p, which is defined as the depth into the material at which the power flux has fallen to $1/e$ of its surface value [27]. When $\varepsilon'' \leq \varepsilon'$, D_p can be approximated according to Eq. (2), where ε' is the dielectric constant and λ_0 is the microwave wavelength.

$$D_p \approx \frac{\lambda_0 \sqrt{\varepsilon'}}{2\pi \varepsilon''}. \quad (2)$$

It is clear from Eq. (2) that the penetration depth decreases as the dielectric loss of the material increases [27]. The practical implication of this is that microwaves cannot penetrate very far into materials that absorb microwaves strongly, and this limits the reactor dimensions.

For the purposes of this work, the process inputs varied were power input, treatment time and coal type. The aim was to determine the effect of these variables on coke quality. In order to optimise the process in terms of energy requirements, the E term in Eq. (1) would need to be manipulated, and this would be done by optimising the cavity design. The information presented here would be vital developing the process further. It is likely that the energy requirements would be dramatically reduced with the development of an optimised microwave cavity. Such a cavity would need to be designed specifically for coke manufacture, and this will only be possible once the key parameters that govern the coking process are fully understood.

2. Materials and methods

2.1. Coal samples

The microwave coking of four different coals was investigated: a high volatile English bituminous coal (Coal A), a low rank semi-bituminous coal from Kaltim Prima (Coal B), a medium rank bituminous coal from Wales, UK (Coal C) and a semi anthracite from Wales, UK (Coal D). The petrographic characteristics and proximate analysis of Coals A–D are given in Table 1.

The coal samples were passed through a jaw crusher and sieved to a size fraction of 1–3.35 mm prior to microwave heating.

2.2. Microwave experiments

The experimental setup is shown in Fig. 1. All experiments used a variable power 6 kW microwave system (HF Dielectric Heater FDU 543VD-02) operating at a frequency of 2.45 GHz with a cylindrical cavity, waveguide and manual three stub tuner for impedance matching purposes. The reflected power was maintained below 10% during the majority of the operating time. Each experiment used 220 g of coal held in a Pyrex beaker covered by glass wool. The glass wool acted as a dielectric barrier to protect the cavity from arcing and to contain the beaker in the event that it cracked during the experiment. All flanges were tightly sealed and the cavity was flushed with 15 L/min N_2 for 15 min (i.e. approximately six reactor volumes) prior to the experiments. The N_2 flow was reduced to 10 L/min during operation, and this maintained the inert environment as well as acting as a carrier gas to remove products that were evolved during processing.

Coke samples were prepared from Coal A using a range of microwave powers (750 W–4.5 kW) and treatment times (15–120 min), representing a range of total energy inputs of 1700–27,200 kWh/t.