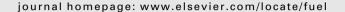


Contents lists available at ScienceDirect

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Direct optical observation of coal particle fragmentation behavior in a drop-tube reactor



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HIGHLIGHTS

- A drop-tube reactor is presented enabling in-situ observation of heated particles.
- Video recordings are provided using a high-speed camera.
- Investigated brown coal particles did not show any fragmentation.
- Fragmentation was observed for a high-volatile bituminous coal and an anthracite.
- Fragmentation will increase with particle size, temperature and residence time.

ARTICLE INFO

Article history: Received 6 August 2015 Received in revised form 29 October 2015 Accepted 3 November 2015 Available online 6 November 2015

Keywords:
Coal
Devolatilisation
In-situ observation
Particle heating rate
Particle residence time
Primary fragmentation

ABSTRACT

An experimental plant - called Single Particle Disintegrator (SPaltor) - is presented that allows an observation of coal particle fragmentation during the thermochemical conversion process. The SPaltor is a labscale drop-tube reactor which runs at atmospheric pressure and temperatures up to 1600 °C. A highspeed camera, a long-distance microscope, and a high-power LED outside the reactor in combination with two mirrors arranged inside the reactor in a 45°-position are used to monitor the inside of the reactor. In this setup, the particle behavior of three different feedstocks were recorded at nitrogen atmosphere. Two recordings are presented in this paper for a reactor temperature of 1400 °C. While a brown coal particle shows no fragmentation, but a bright gas tail due to devolatilisation, an anthracite particle disintegrates into smaller fragments in the first third of the heated zone. For detailed investigation of primary fragmentation, three non-swelling feedstocks were used: A brown coal from Germany, a high-volatile bituminous coal from South Africa, and an anthracite from Germany. Two key figures were used for the description of fragmentation behavior: Fragmentation probability and fragmentation number. The influences of temperature (800–1400 °C), particle size (0.8–3.15 mm), reactor height (0–90 cm), and calculated residence time (0.25-0.5 s) were investigated. Increasing these parameters led to an increase of fragmentation of the anthracite and the high-volatile bituminous coal. In contrast, fragmentation was never observed for the brown coal in any experiment. For the chosen particle sizes, the residence time in the heated zone is smaller than 0.5 s and comparable to free-fall conditions. The heating rate referring to the particle center varies heavily with particle size and coal type.

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1. Introduction

Particle fragmentation is an important phenomenon in coal conversion processes like pyrolysis, gasification, and combustion, esp. during fast heating up and reaction. The change in particle size influences the heat and mass transfer as well as the reaction conditions. The generation of smaller particles through fragmentation processes could also lead to an increased pressure drop in

fixed-bed, to a lower particle residence time in fluidized bed, and a higher conversion rate in entrained flow processes. There are different types of fragmentation which have to be distinguished (Fig. 1).

When a coal particle is heated rapidly in an inert gas atmosphere, primary fragmentation can take place as a result of devolatilization or thermal shock. Devolatilization of volatile matter leads to a rapid pressure increase inside a particle as well as compressive and tensile stresses. Thermal shock can occur due to a temperature gradient inside the particle which arises trough limited heat transfer inside the particle [2]. In reactive gas atmospheres containing O_2 , O_2 , or

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Nomenclature Α front surface (m²) S fragmentation probability (%) thermal diffusivity (m²/s) T_N standard temperature (°C) drag coefficient (-) process temperature (°C) c_D T_0 specific heat capacity of coal particle (J/(kg K)) gas residence time (s) C_P t_{gas} particle residence time (s) D inner diameter of SPaltor (m) tparticle particle diameter (m) gas/particle velocity at 0.9 m (m/s) А $u_{0.9m}$ F_b buoyant force (N) V_P particle volume (m³) drag force (N) \dot{V}_N F_d standard volume flow rate (m³/s) gravity force (N) kinematic viscosity of fluid (m²/s) F_g v_F F_r resulting force (N) χ particle trajectory (m) standard acceleration of free fall (m/s2) χ particle velocity (m/s) g particle acceleration (m/s²) heating rate (K/s) ÿ h I length of SPaltor (m) X coordinate of image matrix (-) m_F fluid mass (kg) coordinate of image matrix (-) particle mass (kg) m_P fragmentation number (-) N **Abbreviations** N_{frag} number of fragmented particles (-) Ash Ash content (proximate analysis) N_{in} number of inserted particles (-) ar as received N_{out} number of obtained particles (-) C Carbon content (ultimate analysis) Nu Nusselt number Fixed Carbon content (proximate analysis) C_{fix} thermal conductivity (W/(m K)) db dry basis Pr Prandtl number Н Hydrogen content (ultimate analysis) particle radius (m) 0 Oxygen content (ultimate analysis) Re Reynolds number (-) N Nitrogen content (ultimate analysis) fluid density (kg/m³) O_F Combustible sulfur content (ultimate analysis) bulk coal particle density (kg/m³⁾ ρ_P VM Volatile matter content (proximate analysis)

H₂O, secondary fragmentation can take place. Heterogeneous gassolid reactions lead to a weakening and break-up of carbon bridges. In the late stage of carbon conversion, many fines will be produced through pore enlargement and coalescence. This process is called percolation. Attrition is a phenomenon which takes place over the entire range of high-temperature processes due to particle-particle-contact or contact between particles and reactor wall and reactor fixtures, respectively. In comparison to primary or secondary fragmentation the generated particles are very small compared to the mother particle [3].

Chirone et al. [4] gave a first literature review about coal particle fragmentation. In the past, most emphasis was put on investigating primary and secondary fragmentation behavior in fluidized bed processes due to the fact that residence time and particle size are associated [2,5–10]. Besides coal particles, also the fragmentation behavior of biomass [11] and inert bed materials like quartzite [12] and limestone [13] was investigated. Only limited information is available about coal particle fragmentation behavior in drop-tube furnaces [14,15].

Senneca et al. [16] developed a "heated strip reactor" that permitted very high heating rates up to 10^4 K/s. Using a near infrared fast camera with a maximum of 38,000 frames/s, they were able to detect the temperature profile of the strip and the particle. Particle counting and particle sizing were done by laser granulometry. The heated strip reactor was further advanced to run at pressures up to 20 bar. Another utilized fixed bed application was a TGA [17].

Attention was also paid to the investigation of mechanical properties which is considered to be an important parameter to describe coal particle fragmentation. Zhong et al. [18] investigated the compressive strength of single brown coal particles. They determined that compressive strength of coal particles increases with decreasing particle size and the utilization of thermal treatment by coal pyrolysis.

Kim et al. [19] observed the ignition behavior of pulverized coal particles using a flat flame burner at high heating rates ($>10^5$ K/s).

They used a high-speed camera to capture the ignition process. In these studies, they observed fragmentation for anthracite and a lignite coal prior to ignition.

In this paper, a new reactor with the possibility of a direct particle observation is presented. The influences of reactor temperature, particle size and residence time on coal particle fragmentation are analyzed and discussed.

2. Experimental

2.1. SPaltor – Single particle disintegrator

A lab-scale experimental plant – called Single Particle Disintegrator (SPaltor) – was planned and built in cooperation with HTM Reetz GmbH. The SPaltor consists of several parts: a single particle dosing system, a heated drop-tube reactor, two mirrors, a high-speed camera, a long-distance microscope, a high-powered LED, an Ulbricht sphere, and a collection container. A simplified scheme of the experimental plant including three pictures of the equipment is presented in Fig. 2.

The drop-tube reactor consists of an Al_2O_3 -tube with an inner diameter of 40 mm. The tube is electrical heated by three heating zones controlled independently over 1 m. The reactor runs at atmospheric pressure and a temperature up to $1600\,^{\circ}$ C. To prevent air inlet, the reactor is flushed with nitrogen of 1 l/min (STP) permanently. The coal particles are inserted one by one via a ball valve on the top of the reactor.

At the upper and the lower part of the reactor, two mirrors are integrated in a 45° position to observe the fragmentation behavior of single coal particles via a combination of high-speed camera (Optronis) and long-distance microscope (Infinity). The highest frame rate of the high-speed camera is 1000 frames/s at the highest resolution of 1280×1024 . All parameters of the camera are listed in Table 1.

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