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Research Paper

Experiment and modeling of low-concentration methane catalytic combustion in a fluidized bed reactor

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HIGHLIGHTS

- The catalytic combustion of 0.15~3 vol. % low concentration methane in a fluidized bed was studied.
- A mathematical model was proposed on the basis of gas–solid flow theory.
- A comparative analysis of the established model with plug flow, mixed flow and K-L models was carried out.
- The axial methane profile along fluidized bed was predicted by using the mathematical model.
- The bed temperature has greater impact on methane conversion than fluidized velocity.

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ABSTRACT

This study undertakes a theoretical analysis and an experimental investigation into the characteristics of low-concentration methane catalytic combustion in a bubbling fluidized bed reactor using 0.5 wt.% Pd/Al_2O_3 as catalytic particles. A mathematical model is established based on gas–solid flow theory and is used to study the effects of bed temperature and fluidized velocity on methane catalytic combustion, and predict the dimensionless methane concentration axial profile in reactor. It is shown that methane conversion increases with bed temperature, but decreases with increasing fluidized velocity. These theoretical results are found to correlate well with the experimental measurement, with a deviation within 5%. A comparative analysis of the developed model with plug flow, mixed flow and K-L models is also carried out, and this further verifies that the established model better reflects the characteristics of low-concentration methane catalytic combustion in a bubbling fluidized bed. Using this reaction model, it was found that the difference in methane conversion between dense and freeboard zones gradually increases with bed temperature; the dense zone reaction levels off at 650 °C, thereby minimizing the difference between the dense and freeboard regions to around 15%. With an increase in bed temperature, the dimensionless methane concentration in the dense zone decreases exponentially, while in the splash zone, it varies from an exponential decay to a slow decrease. In contrast, there is very little change in the homogeneous zone, meaning that combustion mainly occurs in the dense zone and moves toward the lower part of the bed with increasing bed temperature.

1. Introduction

It is common knowledge that low-concentration methane is prevalent in coal bed methane and exhaust gases; its concentration varies depending on the operating conditions [1,2]. However, its low calorific value makes it difficult to use, and therefore, most untreated low-concentration methane is simply released to the atmosphere. This not only causes air pollution, but also represents a colossal waste of energy resources [3,4], thus making it important to find an efficient technique for utilizing low-concentration methane.

Fluidized bed catalytic combustion has been widely used for the combustion of inferior grade fuels because of its wide scope of application, high heat capacity, low light-off temperature and high oxidizing speed [5–11]. It has also been found that low-concentration methane can be oxidized completely at low temperatures in fluidized beds if suitable catalysts are used [12,13]. This fluidized bed combustion of low-concentration methane involves multi-step surface reactions, which in the presence of an excess of oxygen, can be broadly expressed by:

$$CH_4 + 2O_2 \rightarrow CO_2 + 2H_2O \quad \Delta H^0_{298} = -802\, kJ/mol \tag{1}$$

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Earlier research into gaseous fuel fluidized combustion has covered some aspects of fuel–air mixture burning with inert and catalytic particles employed as bed materials, from which some interesting conclusions were reached. Constantineau et al. [14] established a fluidized bed reactor model to simulate the transition from bubbling to slugging in response to changing operating variables such as the superficial gas velocity, bed inventory and bed height. Though this gave a good representation of gas conversion within the slugging and bubbling regimes, it was developed based on a cold test that was used to develop mathematical models for the fluidized bed combustion of gases. Zukowski [15,16] investigated the combustion of premixed natural gas and air mixtures in a bubbling fluidized bed using inert particles as the bed material, through which it was found that the combustion region depends on the average bed temperature. That is, when the temperature is low, combustion mainly occurs in freeboard, but the process moves closer to the distributor at higher bed temperatures. Taking chemical reaction kinetics into account, a mathematical model was subsequently developed to predict the height above the distributor at which the bubbles ignite and explode. In this model, it was assumed that combustion takes place inside bubbles of premixed gas as they move through the bed, and that the bubbles were non-isothermal. Ribeiro and Pinho [17] developed a model for the combustion of methane–air mixtures in fluidized beds that was based on bubble passage and coalescence. By considering the heat transfer between bubble and emulsion phases, as well as the variation in gas flow rate, radiation, and thermal conduction between the fluidized bed and reactor wall, the maximum deviation between bubble and emulsion phases was found to be less than 100 °C. Yang et al. [18] studied the characteristics of low-concentration methane catalytic combustion in a bubbling fluidized bed in which 0.5 wt.% Pd/Al_2O_3 acted as catalytic particles. A mathematical model was established on the basis of a two-phase model and numerical calculations were introduced, with a comparative analysis of the experimental data revealing the maximum deviation of methane conversion to be about 15%.

From a review of the literature, it seems that current research has focused mainly on natural gas, and that any fluidized bed models that have been established were aimed solely at the bed. Clearly, this means that there has been insufficient reaction modeling and investigation into the combustion characteristics of fluidized bed catalytic combustion of low-concentration methane, and therefore, this is something that needs to be studied further. This paper therefore presents a theoretical analysis and experimental research based on a lab-scale fluidized bed reactor supplied with methane–air mixtures, in which 0.5 wt.% Pd/Al_2O_3 was employed as catalytic particles. A mathematical model was established based on the flow characteristics, as well as the reaction and mass balance in the bubbling fluidized bed. Numerical calculations were introduced and compared with experimental data, with the results obtained providing a theoretical basis for low-concentration gas fuel catalytic combustion in fluidized beds.

2. Experimental set up

The experimental system used was primarily composed of a bubbling fluidized bed reactor, electric heater unit, air supply system and temperature control and collection system, all of which are shown schematically in Fig. 1. The reactor consisted of a preheating section ①, gas distributor plate ②, furnace ③, sampling tube ④, cyclone separator ⑤ and cooling water pipe ⑥. The main parameters of this experimental device have been previously published [18], with bed temperature being a particularly important parameter during low-concentration methane catalytic combustion in

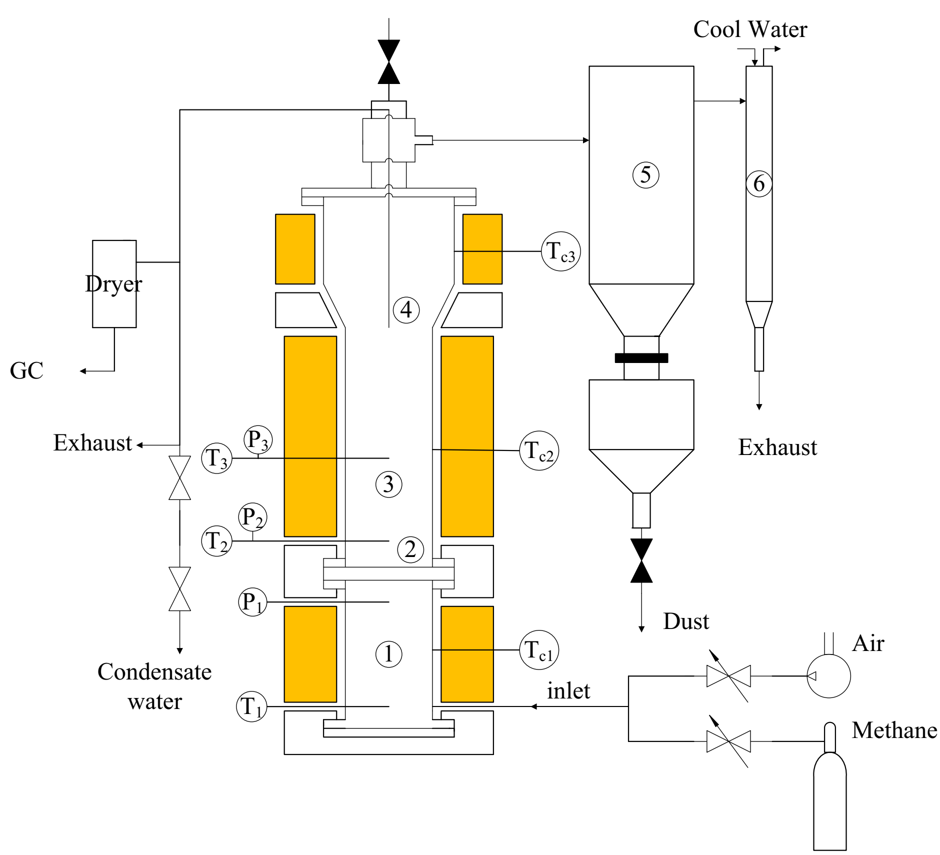

Fig. 1. Schematic diagram of bubbling fluidized bed reactor.