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Experimental studies and the chemical kinetics modelling of oxidation of hydrogen sulfide contained in biogas

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Abstract

This paper presents the results of experimental and numerical research on the process of biogas air combustion. The purpose of the study was to determine the effect of biogas CO_2 content on: (a) variations in SO_2 concentrations in flue gas, (b) variations in the rates of the key reactions of oxidation of H_2S to SO_2.

The subject of investigation were the gas mixtures: CH_4/CO_2 (of 25; 35 and 45 vol % CO_2) with a varying hydrogen sulfide content. The experiments were conducted in a three-zone pipe furnace equipped with a swirl burner (S_g=1.26) with combustion substrate pre-mixing. It was noticed that the consumption of hydrogen sulfide was significantly reduced with the temperature decrease from 1223 K to 1023 K. The increase in the biogas carbon dioxide content inhibited the process of oxidation of H_2S to SO_2.

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Keywords: Hydrogen sulfide; Chemical kinetic modeling; Combustion; Biogas

1. Introduction

For several years now, an increase in the number of biogas plants has been observed in Europe: 14 572 in 2013 [1] and 17 240 in 2014 with a total installed power of 8 293 MW_{el} [2]. On the list of European countries having the most biogas plants – Poland, with 277 plants, is in ninth place. Biogas plants are usually located at waste treatment plants, waste disposal grounds, industrial plants or farms. Depending on the type of raw material used for anaerobic fermentation, the methane content ranges between 50-75 % (v/v). The second main biogas component is carbon dioxide, with a percentage fraction between 50-75 % (v/v). In addition, biogas contains nitrogen, hydrogen sulfide,

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ammonia and hydrogen. Untreated biogas may contain even up to 2% H_2S [3]. The manufacturers of internal combustion engines require a hydrogen sulfide content of $H_2S<50$ ppm [4], while feeding biogas to the natural gas network is allowable at $H_2S<4$ ppm [4]. Even traces of hydrogen sulfide can cause the corrosion of pipelines, compressors or biogas tanks.

The generation of heat and electric energy from the combustion of biogas can be realized based on the following technologies: biogas flameless combustion [5], oxy-fuel combustion [6], hydrogen-enriched biogas combustion [7], combustion in gas turbines [8], combustion in HCCI engines [9] and swirl combustion [10], in spite of existing limitations, such as the low mean calorific value of biogas, amounting to 21.5 MJ/m^3 [5], and the high cost of biogas purification and combustion installation modernization (burner replacement). The process of biogas combustible component oxidation depends on the applied combustion technology, combustion temperature and the oxidizer fraction of the combustible mixture. For the whole hydrogen sulfide contained in biogas be able to be oxidized to SO_2, a combustion temperature of $1000\ K<T<1473\ K$ is required [8,11]. If combustion temperature is too low, the flame lacks of radicals, such as SH, HSS and H, and the H_2S will be oxidized only partially. Equivalence ratio (Φ) of the combustible mixture depends on the biogas and combustion technology. The use of the equivalence ratio $\Phi \ll 1.0$, allows unburned hydrocarbons and CO occurring in the combustion products to be limited.

2. Experimental Section

The experiments were conducted for the following gas mixtures: CH_4/CO_2 (of 25; 35 and 45 vol % CO_2) with a varying hydrogen sulfide content. The varying H_2S content of the combustible mixtures tested resulted from the equivalence ratio preset as per Table 1, because synthetic cylinder air contained 15 ppm H_2S. The lower the equivalence ratio value, the more H_2S is contained in the combustible mixture. The H_2S concentration lay in the range of 131-268 ppm. The choice of the hydrogen sulfide concentration range of H_2S < 300 ppm resulted from the experience of researchers [12-14] dealing with the oxidation of H_2S. Low temperature in the combustion reactor, an insufficient amount of oxygen and an increased H_2S content of the combustible mixture, all favour the formation of elementary sulphur. Clogging of the flue gas sucking probe with elementary sulphur is possible in that case. In this investigation, sulphur deposition on the ceramic flue gas suction probe was observed after two days of experiments.

Table 1. H_2S content in biogas (ppm).

Φ	25 %CO_2	35 %CO_2	45 %CO_2
0.4	267.9	232	195.5
0.45	237.9	206	174.4
0.5	214.3	185.7	157.2
0.55	195	169	142.9
0.6	179	155	131.2

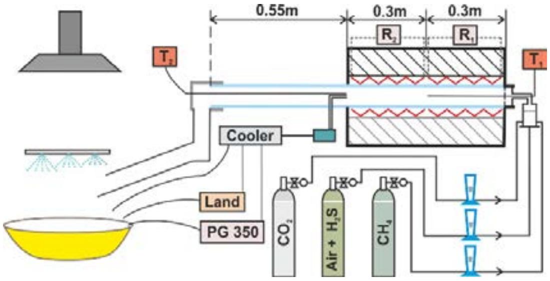

Fig. 1. A schematic diagram of the test stand.

The experiments were conducted on a test stand, as illustrated schematically in Fig. 1. The main part of the test stand is a quartz pipe furnace chamber (having two heating zones and one cooling zone) equipped with a swirl burner with a combustion substrate mixing capability. The design of the burner with a geometric swirl number of $S_g=1.26$ is described in detail in reference [15]. For preset temperatures of 1223 and 1423 K (in the first and the second furnace zone, respectively) and for lean fuel conditions (an equivalence ratio of $0.4\leq \Phi \leq 0.6$), the following gas concentrations were measured: H_2S – with a LAND Lancom Series II analyzer; and CO, CO_2 and SO_2 – with a Horiba PG350 analyzer, in the dry flue gas leaving the second furnace zone. The flue gas was sucked in to the gas cooler with a ceramic probe, and then directed to respective analyzers. To inhibit reactions occurring inside the probe sucking the flue gas at a temperature of approx. 1223 and 1423 K, water cooling could be employed. However, due to the small size of the probe and the test stand there was a risk that an excessive cooling of the flue gas could cause the