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Flammability limits and burning characteristics of CO–H₂–H₂O–CO₂–N₂ mixtures at elevated temperatures

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ABSTRACT

The paper presents the results of an experimental evaluation of the flammability limits and maximum pressure rise rate for multi-component (CO–H₂)–O₂–(H₂O–CO₂–N₂)-mixtures at elevated temperatures of 170 °C and 250 °C at ambient pressure of 1 bar. A spherical explosion chamber with a volume of 8.2 dm³ was used for the experiments. A pressure method and a high-speed camera combined with a schlieren system for flame visualization were used in this work. Flammability limits and pressure rise rates are presented for multi-component mixtures with a steam concentration of 60% and a wide variation of fuel and oxidizer concentrations in which the H₂ concentrations was limited to 3%.

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Introduction

The study of the burning characteristics of multi-component CO–H₂–O₂–H₂O mixtures is very important from the practical point of view. High temperature reaction processes in industry, core melt accidents in nuclear power plant or large industrial fire events are able to produce large amounts of binary fuel mixtures consisting of carbon monoxide and hydrogen. An example is the gas generated in waste gas treatment systems of steel manufacturing plants [1], in which

a high temperature gas mixture of H₂, O₂, CO, CO₂, N₂ and inert dust particles are present. Very high initial temperature of this mixtures is first reduced by injection of cold water (in an evaporation cooler) and so larger dust particles are collected inside the evaporation cooler. To separate very fine dust particles from the gas phase behind the evaporation cooler an electrostatic precipitator [ESP] is a probate and efficient method. Typical feed temperatures for an ESP lie in the range from 170 °C to 250 °C, so that the multi component gas mixture has a high steam content. During normal operation conditions the presence of combustible mixtures is excluded

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by assuring an oxygen deficit in the process gas. In case of a severe process disturbance reactive CO–H₂–O₂ mixtures could develop inside the ESP and, due to the high voltage used for charging dust particles, an ignition cannot be excluded. For the safety design of an ESP vessel and an effective vessel venting system the knowledge of the flammability limits and maximum pressure rise rates of the multi component gas mixture at elevated temperatures are necessary. Flammability limits of hydrogen–carbon monoxide–air mixtures from 20 to 300 °C were measured in Ref. [2]. In few cases up to 20 vol. % steam were used as diluent. Lean flammability limits of CO–H₂–air mixtures at 25 °C initial temperature were measured in Ref. [3]. Flammability limits of hydrogen enriched natural gas are presented in Ref. [4]. The effects of CO₂ and H₂O dilution on laminar burning characteristics of CO/H₂/air mixtures were investigated in Ref. [5] and another work concentrates on laminar burning velocity [6]. In contrast to CO–H₂–mixtures the study of H₂/hydrocarbons–mixtures is in an advanced state, an overview of the investigations of hydrogen enriched hydrocarbons is given in Ref. [7]. For O₂, CO, CO₂, N₂ and H₂ with a high steam content no data on flammability limits and burning characteristics are available [1]. The goal of this work is the experimental investigation of the important combustion characteristics of (CO + H₂)–O₂–(H₂O + CO₂+N₂) gas mixtures at temperatures of 170–250 °C and atmospheric pressure conditions in a spherical explosion chamber. The range of the studied mixture compositions is listed in Table 1.

Experiments

Experimental facility and gas injection system

The experiments to investigate the flammability limits and pressure rise rates of multi-component mixtures were performed in a spherical stainless steel explosion bomb of 25 cm internal diameter and an inner volume of $V = 8.2 \text{ dm}^3$ (Fig. 1), for further details see also [8] and [9]. The bomb has two quartz windows for optical observations as well as ports for two pressure gauges (PCB Type A36 and A22) and NiCr/Ni thermocouples to record pressure and temperature during the combustion process. The ignition via a high voltage (60 kV) high frequency (20 kHz) spark is positioned in the centre of the explosion bomb by using two electrodes. Due to the design of the facility the electrodes meet in an angle of 90°. To keep a centreignition with large gap size the tips of the electrodes were shifted up and down. Shape and size of the bomb were

chosen in accordance with the German Standard PR EN1839 (B) for flammability tests. The schematic of the test facility is shown in Fig. 1. The facility consists of the heated explosion bomb and a dynamic gas mixing and gas injection systems. The metering of the gas components was performed by mass flow controllers, the purities of the supply gases and the specifications of the flow controllers are given in Table 2. For the metering of the steam component a mass flow controller for liquid water was used. The constant liquid water flow was evaporated in a steam generator afterwards. The fuel and oxidizer components were first mixed separately in a small dynamic mixing chamber under ambient temperature conditions, while the inert gas components were mixed under heated conditions in a separate dynamic mixing chamber. Both gas flows join each other near the final dynamic mixing chamber. The accuracy of the flow controllers was verified for dry mixtures by a multi-component (H₂, O₂, CO, CO₂) gas analyser for continuous flows. Therefore all possible binary, ternary and quaternary mixture variations with and without N₂ fraction were checked. The ratio of the steam flow to the dry mixture flow was verified using the ratio of the linear pressure increases during the filling procedure of the 9 dm³ vessel with dry mixtures and the filling procedure with mixtures with a steam concentration of 60 vol. %. Explosion bomb, steam generator, inlet gas lines and valves were heated with electrical heating cables. The temperature of the explosion bomb was additionally controlled via NiCr/Ni thermocouples which were placed close to the quartz glass windows inside the metal body of the bomb. Two collimated NiCr/Ni thermocouples measured the gas temperature in different distances to the wall (5 mm and 50 mm) inside the explosion bomb. In selected experiments the optical access to the explosion bomb was used to capture the ignition process by using high speed shadowgraphy.

Experimental procedure

To prepare the multi-component mixture in the best possible way, the steam flow rate was fixed to 400 g/h (8.32 STP/dm³ per minute) and also the steam concentration of the initial mixtures was fixed to 60 vol. % in all experiments. So the total flow rate of the studied mixtures was 13.86 STP/dm³ per minute in all experiments. The concentrations of fuel (H₂, CO) and O₂ were selected to be together less than 40 vol. % of the mixture. In the cases where fuel and oxidizer were less than 40 vol. % of the mixture, the inert gases CO₂ and N₂ were added in equal portions to reach a total feed flow of 13.86 STP dm³ per minute. An additional 9 dm³ vessel was used to damp the start-up of the filing process of the spherical combustion chamber (Fig. 1).

Ignition device

The ignition device and its energy release is an important factor for the measurement of flammability limits. In this work an ignition device which produces a high voltage (60 kV) and high frequency (20 kHz) spark between two electrodes in the centre of the explosion bomb was used. To estimate the energy release of the ignition device, the pressure increase due to the thermal energy release inside the explosion chamber which is only driven by the spark was measured. In

Table 1 – Investigated gas concentration ranges.

Component	Concentration range at 170 °C	Concentration range at 250 °C
CO	0 – 20 vol. %	6 – 20 vol. %
H ₂	0 – 3 vol. %	0 – 3 vol. %
O ₂	3 – 20 vol. %	3 – 20 vol. %
H ₂ O	60 vol. % in all tests	60 vol. % in all tests
CO ₂	0 – 12 vol. %	1 – 11 vol. %
N ₂	0 – 11 vol. %	1 – 11 vol. %

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