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Mechanisms of high-pressure hydrogen gas self-ignition in tubes

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Abstract

This paper describes a numerical and experimental investigation of hydrogen self-ignition occurring as a result of the formation of a shock wave. The shock wave is formed in front of high-pressure hydrogen gas propagating in a tube. The ignition of the hydrogen–air mixture occurs at the contact surface of the hydrogen and oxidant mixture and is due to the temperature increase produced as a result of the shock wave. The required condition for self-ignition is to maintain the high temperature in the mixture for a time long enough for inflammation to take place. The experimental technique employed was based on a high-pressure chamber pressurized with hydrogen, to the point of a burst disk operating to discharge pressurized hydrogen into a tube of cylindrical or rectangular cross section containing air. A physicochemical model involving gas-dynamic transport of a viscous gas, detailed kinetics of hydrogen oxidation and heat exchange in the laminar approach was used for calculations of high-pressure hydrogen self-ignition. The reservoir pressure range, when a shock wave is formed in the air that has sufficient intensity to produce self-ignition of the hydrogen–air mixture, is found. An analysis of governing physical phenomena based on the experimental and numerical results of the initial conditions (the hydrogen pressure inside the vessel, and the shape of the tube in which the hydrogen was discharged) and physical mechanisms that lead to combustion is presented.

Keywords: Self-ignition; Hydrogen; Shock wave; Tubes

1. Introduction

The interest in hydrogen as an energy carrier has led to a number of studies of hydrogen safety issues, with a view to the development of new relevant safety codes and standards. Although a promising field, hydrogen energy development brings up safety problems relating to hydrogen storage and processing, consumption, and transportation. One particularly important aspect of this today is the safety of high-pressure storage reservoirs. Compressed hydrogen storages at pressures up to 700 atm are under consideration by the transportation industry.

One particular problem relating to pressurized hydrogen storage, which has a number of times over the last century, is high-pressure hydrogen leaks igniting for no apparent reason. Various hypotheses have been put forward to attempts to describe these ignitions. There have been several theories: the reverse Joule–Thompson effect, the electrostatic charge generation, the diffusion–ignition, the sudden adiabatic compression, and the hot surface ignition (Astbury & Hawksworth, 2005). Edeskuty and Stewart (1996) reported the possibility of shock wave ignition of premixed hydrogen–air mixtures omitting the question how the shock wave may interact with premixed mixture.

The diffusion–ignition processes of unmixed flammable gas–air was postulated by Wolański and Wójcicki (1973), who demonstrated that ignition occurred, when hydrogen under high pressure was submitted to a shock wave in a tube filled with air or oxygen. The ignition occurred even when the surrounding temperature was below the hydrogen auto-ignition threshold and, was a result of a sharp jump in the temperature of the combustible mixture caused by diffusion, when the hydrogen surface came into contact with the surrounding oxidizer heated by the primary shock wave. Baev, Shumskii, and Yaroslavtsev (1983), Baev, Buzukov, and Shumskii (2000) investigated self-ignition of hydrogen when it came into contact with the surrounding

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air. In difference with the experiment of Wolański and Wójcicki (1973), they heated hydrogen at room temperature without any shock up to 575 K then released it into a partly closed tube. Shock wave reflection from the partly closed end resulted in hydrogen self-ignition.

In 2005, three papers (Bityurin, Bocharov, & Filimonova, 2005; Bazhenova, Bragin, Golub, Scherbak, & Volodin, 2005; Liu, Tsuboi, Sato, Higashino, & Hayashi, 2005) were published devoted to the numerical investigation on the possibility of diffusion–ignition for particular cases of where hydrogen came into contact with an oxidizer. Liu et al. (2006) made a numerical analysis of auto-ignition in high-pressure hydrogen jets coming into contact with air and proved that it was possible.

In the paper of Mogi, Shiina, Kim, and Horigushi (2006), a rupture disk was used in the rapid discharge of high-pressure hydrogen into a tube 5–10 mm in diameter, 3–185 mm in length, with an open end. The failure pressure was varied from 40 to 400 bar. Ignition of the hydrogen jet was observed in the extension tube.

In the paper, self-ignition of high-pressure hydrogen releases into tubes of cylindrical and rectangular cross sections is investigated both experimentally and numerically. The initial conditions (the hydrogen pressure in the vessel and the length of tube for the two tube cross-section shapes), which lead to ignition are analyzed based on numerical simulation and an experimental investigation of the reacting gas discharge. Mechanisms leading to hydrogen self-ignition in the tube are determined.

A recent publication of Golub et al. (2007) demonstrates the numerical solution of the hydrogen self-ignition when hydrogen is released into a semi-confined space. The paper includes an experimental investigation of impulse jet structure, numerical simulations of hydrogen mixing with air and hydrogen jet ignition. Diffusion self-ignition mechanism occurs and appears to be responsible for hydrogen jet ignition.

The paper of Dryer et al. (2007) demonstrates the "spontaneous ignition" (auto-ignition/inflammation and sustained diffusive combustion) from sudden compressed hydrogen releases, for which little fundamental explanation, discussion, or research foundation exists, and which is apparently not encompassed in recent formulations of safety codes and standards for piping, storage, and use of high-pressure compressed gas systems handling hydrogen. Both experiments and simple transient shock theory along with chemical kinetic ignition calculations are used to support interpretation of observations and qualitatively identify controlling gas properties and geometrical parameters. Similar considerations for compressed methane, mixtures of light hydrocarbons and methane (simulating natural gas), and larger carbon number hydrocarbons show that similar auto-ignition phenomena may occur with highly compressed methane or natural gas but are unlikely with higher carbon number cases, unless the compressed source and/or surrounding air is sufficiently pre-heated above the ambient temperature. Spontaneous ignition of compressed hydrocarbon gases is also generally less likely, given the much lower turbulent blow-off velocity of hydrocarbons in comparison to that for hydrogen.

The aim of this investigation is the numerical and experimental determination of governing physical phenomena affecting the hydrogen self-ignition at the discharge into the tube filled with air.

2. Experimental investigation of hydrogen self-ignition

An experimental investigation of hydrogen self-ignition in air was carried out in tubes of cylindrical and rectangular cross sections of lengths 65–185 mm and cross-sectional areas of 20 mm^2. The experimental setups are presented in Fig. 1 and consist of a hydrogen cylinder (1) equipped with a valve and a manometer (2) for measuring the pressure in the high-pressure hydrogen chamber (3), a diaphragm block (4,5) connected to the chamber from one side and the low-pressure chamber (cylindrical tube or rectangular tube) (8) from the other side. The tube is connected to the buster chamber (9). The high-pressure chamber is filled with hydrogen from the cylinder (1). The high-pressure chamber and the tube are separated with a copper burst disk of 0.1–0.2 mm in thickness. As the pressure in the reservoir reaches the critical value the burst disk fails and hydrogen discharges into the tube (8) forming the leading shock wave. Three pressure transducers PCB 113A24 (6) and light sensors

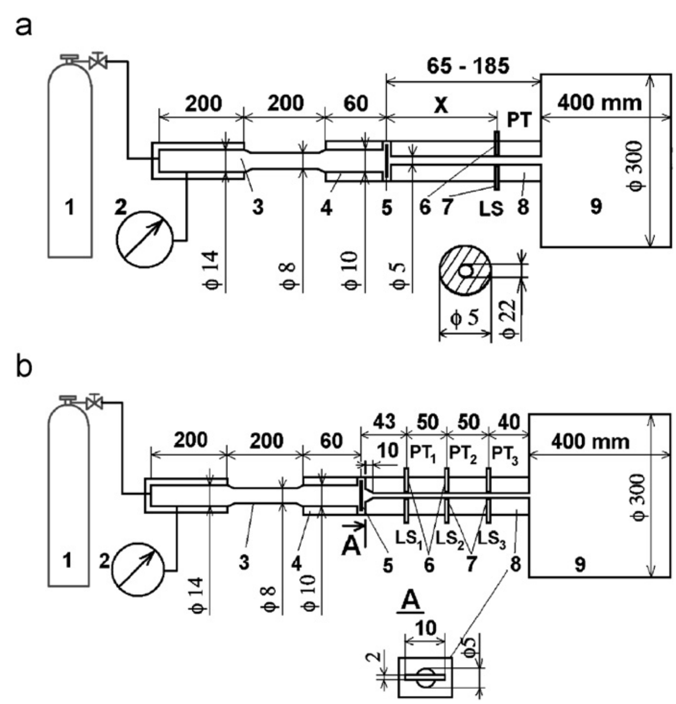

Fig. 1. Schematic of experimental setups. (a) low pressure tube with cylindrical cross section; (b) low pressure tube with rectangular cross section. 1—hydrogen cylinder, 2—manometer, 3—high pressure chamber, 4—burst disk block, 5—copper burst disk, 6—pressure transducers (PT), 7—light sensors (LS), 8—low pressure chamber (tube); 9—buster chamber. *X*—distance between burst disk and pressure transducer.