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Experimental study of minimum ignition energy of lean H_2-N_2O mixtures

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Abstract

Ignition energies for short duration (<50 ns) spark discharges were measured for undiluted and nitrogen-diluted H_2-N_2O mixtures of equivalence ratios $\phi = 0.15$ and 0.2, dilution of 0% and 20% N_2, and initial pressures of 15–25 kPa. The ignition events were analyzed using statistical tools and the probability of ignition versus spark energy density (spark energy divided by the spark length) was obtained. The simple cylindrical ignition kernel model was compared against the results from the present study. Initial pressure has a significant effect on the width of the probability distribution, ranging from a broad ($P = 15$ kPa) to a narrow ($P = 25$ kPa) probability distribution indicating that the statistical variation of median spark energy density increases as initial pressure of the mixture decreases. A change in the equivalence ratio from 0.15 to 0.2 had a small effect on the median spark energy density. The addition of 20% N_2 dilution caused a significant increase in the median spark energy density when compared to no dilution. The extrapolation of the present results to atmospheric pressure, stoichiometric H_2-N_2O indicates that the electrostatic discharge ignition hazards are comparable to or greater than H_2-O_2 mixtures.

Keywords: Spark ignition; Minimum ignition energy; Hydrogen; Nitrous oxide; Nuclear waste safety

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1. Introduction

The Hanford site, located in south central Washington, has been used for more than 40 years to produce plutonium for the United States' nuclear weapons arsenal. This led to the largest amount of localized nuclear waste in the United States. Since plutonium production in the United States ceased in the late 1980s, the Hanford site has been engaged in a waste cleanup phase that will extend several more decades [1,2]. The stored waste at the facility continuously generates gaseous compounds that form mixtures of H_2, nitrous oxide (N_2O), O_2, N_2, ammonia (NH_3) and methane (CH_4) [3]. The hazard related to the accidental ignition of these gas mixtures must therefore be considered, particularly that of hydrogen–nitrous oxide (H_2-N_2O) mixtures.

Over the past 20 years, numerous studies have been conducted to characterize the explosive properties of H_2-N_2O. Pfahl et al. studied the flammability limits of H_2-N_2O mixtures [4]. The flame speeds of H_2-N_2O-Ar mixtures and H_2-N_2O-N_2 mixtures were investigated by Mével et al. [5] and Bane et al. [6], respectively. Mével et al. [7,8] and Javoy et al. [9] measured ignition

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delay times. Liang et al. [10] studied deflagration to detonation transition in undiluted H_2-N_2O mixtures. Akbar et al. [11] and Kaneshige et al. [12] measured the detonation cell size and Zhang et al. [13] measured the critical energy required for direct detonation initiation. Despite these extensive studies, we are not aware of any data on the electrostatic discharge ignition potential for these mixtures. The objective of the present study is to characterize the energies needed for electrostatic discharge ignition of diluted and undiluted H_2-N_2O mixtures. Experiments were performed for selected equivalence ratios, dilution levels, initial pressures, flanged and plain electrodes, and compared to a simple model of spark ignition.

2. Materials and methods

2.1. Ignition system

We have developed a low-energy (60 µJ–1.8 mJ), short duration (<50 ns) capacitive spark ignition system capable of producing 1.5–4 mm discharges with high repeatability. This simulates the type of spark discharge encountered in typical electrostatic ignition hazards. The circuit consists of a 0–30 kV high voltage power supply connected to a 10 GΩ charging resistor in series with a 3–30 pF or a 15–250 pF variable vacuum capacitor. The capacitor is connected in parallel with the spark gap, so that it discharges through the gap when the capacitor reaches the gap breakdown voltage. A function generator provides a ramp signal to the high voltage power supply allowing it to ramp up from 0 to 10 kV. The ramp time is longer than the maximum capacitor charging time; therefore, the voltage on the capacitor at the time of breakdown can be measured at the output of the power supply. At breakdown, the spark current is measured using a Bergoz fast current transformer and triggers an oscilloscope that digitizes the output at a sampling rate of 2 GS/s. A 5 V output signal on the oscilloscope is then used to trigger a delay generator that triggers a high voltage relay, the data acquisition, and a high-speed camera. The high voltage relay is used to disconnect the capacitor from the high voltage power supply after the spark has occurred to prevent multiple sparks.

2.2. Estimating spark energy

The energy of the spark is approximated as the difference between the initial stored energy in the capacitor and the residual energy [14,15]:

$$E_{spark} \approx E_{stored} - E_{residual}. \tag{1}$$

The stored energy and residual energy are given by

$$E_{stored} = \frac{1}{2} C V^2_{breakdown}, \tag{2}$$

$$E_{residual} = \frac{1}{2} \frac{Q^2_{residual}}{C}. \tag{3}$$

The total capacitance, C, is measured using a Keithley Model 6517A Electrometer/High Resistance Meter and the voltage at breakdown, $V_{\text{breakdown}}$, is measured using a high voltage probe. The residual charge, Q_{residual}, is found by subtracting the charge deposited into the spark channel from the stored charge in the capacitor. The integral of the spark current (equal to the charge deposited into the spark channel) is found by numerically integrating the waveform obtained from the current transformer:

$$Q_{residual} = Q_{stored} - Q_{spark}, \tag{4}$$

$$Q_{residual} = C V_{breakdown} - \int i(t)dt. \tag{5}$$

The uncertainty in the spark energy measurement is calculated given the uncertainty in the voltage (4%), capacitance (3%) and current measurements (0.5%), which leads to an uncertainty of approximately 10% in the computed spark energy.

2.3. Combustion vessel and diagnostics

The ignition experiments were performed in a closed, cylindrical, stainless steel combustion vessel with a volume of approximately 22 L. Two parallel flanges were used to mount the spark gap electrodes, and two additional flanges held windows for visualization. A remotely controlled plumbing system is used to evacuate the chamber and accurately fill it with gases using the method of partial pressures. A Heise 901A manometer with a precise digital readout measures the static pressure so the gases can be filled to within 0.03% of the desired gas pressure, providing precise control over the mixture composition. Three different methods were used for ignition detection. First, the pressure rise from the combustion was measured using a pressure transducer. This measurement also allowed us to determine the peak pressure rise in the vessel. Second, the temperature rise was detected using a K-type thermocouple located inside the vessel. The third method used to detect ignition was schlieren visualization of the flame propagation recorded using a high-speed camera. Two different schlieren systems were used: the first had a large field of view (117 mm diameter) to visualize the flame propagation and the second had a small field of view (5.5 mm diameter) to visualize the early stages of the spark discharge and flame kernel development.