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Detonation in hydrogen–nitrous oxide–diluent mixtures: An experimental and numerical study

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ABSTRACT

Knowledge of H_2–N_2O mixtures explosive properties is important to the safety of nuclear waste storage and semi-conductor manufacturing processes. The present study provides new experimental data on H_2–N_2O detonations, and proposes a thermochemical model which is used to numerically simulate detonation propagation. Detonation cell size has been measured in a variety of H_2–N_2O–Ar mixtures. Even at low initial pressure, these mixtures are very sensitive to detonation with cell size of few millimeters. Using a reduced version of a detailed reaction scheme, 2-D Euler simulations have been used to examine the features of detonation in H_2–N_2O–Diluent mixtures. A PLIF model has been applied to allow for direct comparison with experimental results. Statistical analysis of the cellular cycle dynamics has been performed.

1. Introduction

Hydrogen–nitrous oxide chemistry plays an important role in the analysis of potential hazards in the storage and processing of high-level radioactive waste [1]. It is also important for silane oxidation by nitrous oxide [2,3] because the thermal decomposition of silane is faster than that of nitrous oxide and results in the release of molecular hydrogen [4]. Silane-nitrous oxide mixtures are widely used in the semi-conductor industry [5] in order to form insulator or protective layers [6–9] in a wide variety of applications [10–12]. These mixtures have been involved in some accidental combustion events [13].

Among possible combustion events, detonation is the most severe and has the greatest potential for structural damage. Although direct initiation of detonation is usually considered unlikely, studies have shown that confinement and congestion can lead to detonation initiation via the deflagration-to-detonation transition (DDT) process [14]. The estimation of detonation properties, particularly cell size, is of fundamental importance for evaluation of detonation hazards in industrial processes.

Previous studies on hydrogen–nitrous oxide mixtures have generated data sets on the ignition delay time [15–26], the laminar burning speed [27–35], and the minimum ignition energy [36]. Concerning detonation, Zhang et al. measured the critical energy for direct initiation [37] and the cell sizes have been measured [38,39] in the frame work of nuclear waste storage risk assessment. Although effects of equivalence ratio, initial pressure and dilution have been extensively studied, the available data on the cell size are limited compared to hydrocarbon–air or hydrogen–air mixtures.

The objectives of the present study are: to provide additional experimental data on the detonation cell size in hydrogen–nitrous oxide mixtures; to propose a realistic chemical reaction model validated against shock tube and flow reactor studies; and to carry out numerical simulations of detonation front structure based on a reduced version of the reaction model.

2. Materials and methods

2.1. Experimental set-up

Hydrogen–nitrous oxide–argon mixtures were prepared from high purity grade gases supplied by Air Liquide. Each gas was introduced in a 10 L glass tank using the partial pressure method and mixed by a magnetic stirrer for at least half an hour prior to

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Table 1
Error criteria for the reduction of detailed kinetic schemes.

Parameter	Definition	Error tolerance
Time to peak thermicity	$\delta t_{\sigma_{max}} = \frac{\lvert t_{\sigma_{max}} - t^{ref}_{\sigma_{max}} \rvert}{t^{ref}_{\sigma_{max}}}$	10%
Maximum thermicity	$\delta\sigma_{max} = \frac{\lvert \sigma_{max} - \sigma^{ref}_{max} \rvert}{\sigma^{ref}_{max}}$	10%
Equilibrium conditions	$\Delta T_{eq} = \lvert T_{eq} - T^{ref}_{eq} \rvert$	50 K
	$\delta W_{eq} = \frac{\lvert W_{eq} - W^{ref}_{eq} \rvert}{W^{ref}_{eq}}$	2%
Temporal profiles	$\delta_{max}\sigma = max[\delta\sigma(t_n)]$	15%
	$\Delta_{max}T = max[\Delta T(t_n)]$	150 K
	$\delta_{max}W = max[\delta W(t_n)]$	5%

σ: thermicity, t: time, T: temperature, W: mean molar mass, δ: relative error, Δ: absolute error, *max*: maximum, *eq*: equilibrium, *ref*: reference corresponding to the detailed kinetic scheme.

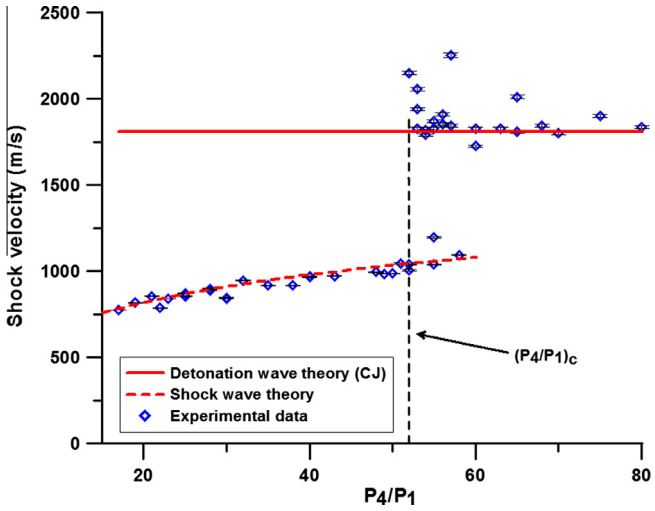

Fig. 1. Shock velocity in H_2–N_2O–Ar mixtures as a function of (P_4/P_1) ratio. Initial conditions: $\Phi = 1$; $X_{Ar} = 0.5$; $P_1 = 10$ kPa; $T_1 = 295$ K.

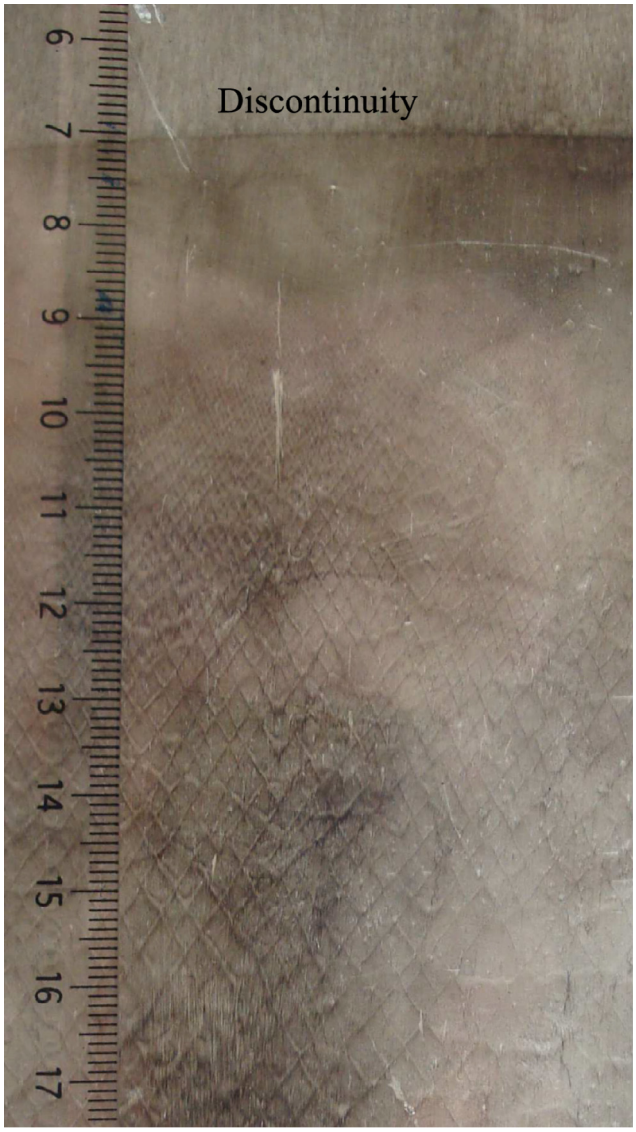

Fig. 2. Experimental soot record of a detonation onset in a H_2–N_2O–Ar mixture. Propagation is from top to bottom. Initial conditions: $\Phi = 2.5$; $X_{Ar} = 0.2$; $P_1 = 10.3$ kPa; $T_1 = 295$ K.

experiments. The initial conditions were varied within the following ranges: equivalence ratio, Φ, between 0.3 and 2.5; dilution from 20 to 60 mol% of Ar; initial pressure between 7 and 35 kPa; and initial temperature of 295 K. Detonations were initiated by incident shock waves using a shock-tube. The shock tube is made of stainless steel with a driver section of 0.9 m long and with an inner diameter of 128 mm. The driven section is either 3.8 or 4.6 m long and has an inner diameter of 78 mm. The two parts of the tube are linked to two vacuum pumps and separated by a double membrane system which allows a good control of the driver section pressure, P_4. Four pressure transducers are mounted flush to the driven section inner wall and allow to measure the shock velocity with an accuracy of 1%. The measured velocity is then used to calculate the temperature and pressure conditions behind the shock wave. The soot record method was used to determine the detonation cell size. The soot foils were located at the end of the driven section. Soot records were digitized and analyzed using the Visilog software.

2.2. Kinetic modeling

The chemical kinetic model used in the present study consists of 203 reactions and 32 species [24,3]. It was mainly based on the Konnov [40] and Mueller et al. [41] mechanisms. The rate constant for the N_2O decomposition reaction was taken from Javoy et al. study [25]. A kinetic sub-set for excited OH radical, OH^*, was based on the mechanisms of Hidaka et al. [21] and Hall and Petersen [42,43]. All thermodynamic data were taken from the Konnov and Mueller et al. models except those for OH^*, which were from Hall and Petersen. Shock tube and flow reactor experiments were both modeled using the SENKIN code [44] from the CHEMKIN II package. Constant volume and constant pressure adiabatic reactor models were chosen to model the experimental conditions of shock tube and flow reactor tests, respectively. For the flow reactor data set, the time-shifting method was used as described by Yetter et al. [45]. Sensitivity and reaction pathway analysis were also performed using the SENKIN code.

2.3. Kinetic scheme reduction

The detailed kinetic model is too large to be directly applied to multidimensional detonation simulations. To allow simulations with the 2-D Euler code described below, the kinetic scheme must