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An experimental investigation of detonation limits in hydrogen–oxygen–argon mixtures

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ABSTRACT

The present paper reports the results of an experimental study of detonation limits for $H_2/O_2/Ar$ mixtures. Two stoichiometric mixtures ($2H_2 + O_2 + 3Ar$ and $2H_2 + O_2$) in three different diameter round tubes ($D = 1.8$, 4.6 and 10.9 mm) were tested. The choice of the mixture represents those considered as "stable" with a regular cellular pattern and "unstable" with an irregular cellular pattern. Detonation velocity was measured by ionization probes spaced at regular intervals along the small tubes. Consistent with previous findings, the present results show that well within the limits the detonation wave in hydrogen mixtures propagates at a steady velocity close to the theoretical Chapman-Jouguet (CJ) value. With decreasing initial pressure, the velocity deficit increases. It is found that the detonation velocity decreases with decreasing tube diameter, which is a result of the wall boundary layer effect being more prominent for smaller tubes. At the limiting pressure, the steady velocity deficit for both tested mixtures in three different diameter tubes is about 15–18%. Velocity deficits were also estimated theoretically using the Fay model. For the mixture of $2H_2 + O_2 + 3Ar$, good agreement is found between the theoretical prediction and the experimental result of detonation velocity. For the mixture of $2H_2 + O_2$, however, the theoretical prediction deviates from the experimental measurement. The latter thus suggests that, apart from losses due to the flow divergence caused by the boundary layer effect, instabilities are also significant for the detonation propagation and failure in $2H_2 + O_2$ mixture. Lastly, at the limits, the value of D/λ is found to agree well with that for the onset of single-headed spin detonation. Thus, it can be concluded that the single-headed spin criterion can also be used for defining the detonation limits for hydrogen mixtures.

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Introduction

The rapid growing worldwide energy needs and greenhouse gas emissions continue to pose great challenges for the existing energy infrastructures. To address these issues, hydrogen has long been proposed as an excellent alternative energy source to replace either gasoline and/or natural gas due to its unique properties, i.e., broad limits range of flammability [1], low ignition energy [2], lack of hydrocarbon and carbon dioxide in combustion products [3]. Indeed, the hydrogen usage is currently increasing and spreading out to many branches of industry including chemical production, metal refining, electronics manufacturing, vehicles and propulsion [4–6]. However, the aforementioned distinct properties of hydrogen may also be considered as disadvantages, and significant safety concerns are associated with hydrogen [7,8]. Because of its wide flammability limit and low ignition energy, a hydrogen cloud could be easily ignited once leakage occurs, generating a slow or fast deflagration, or even a detonation. Although a deflagration is generally the most probable mode of combustion, it could continuously accelerate and undergo an abrupt transition to a detonation under appropriate conditions depending on the concentration, initial pressure, and the confinements involved [9–17,48,49]. Potential risks concerning the occurrence of hydrogen detonation have to be clearly identified and taken into account while establishing safety standards, methodologies and regulations for engineering systems working with hydrogen.

One of the dynamic parameters of detonation wave is the detonability limit which is of significance both from a safety engineering as well as fundamental point of view. Detonation limits refer to the conditions outside of which a steady self-sustained propagation of a detonation wave is not possible [18–20]. The limit phenomenon is a consequence of adverse effect of boundary conditions on the propagation mechanism of a self-sustained detonation. Applying to the industry, detonation limit is equivalent to the conventional parameter of quenching distance or maximum experimental safe gap (MESG) for deflagration used commonly for safety assessment [12]. However, the mechanism of failure is not the same due to different physical processes that controlled their propagation [18]. It has been experimentally well-established that in small diameter tubes as the detonation limits are approached the detonation velocity decreases rapidly [19–22]. The velocity deficit can be attributed to various effects such as the heat and momentum losses to tube walls, flow divergence caused by the boundary layer effect as well as the interference of tube walls with intrinsic instability of the cellular detonation front. Because of these complex interactions, no complete theory for the determination of detonation limits has yet been developed. In addition, near failure of detonation wave, the velocity fluctuations become increasingly large, making the experimental measurement difficult to distinguish precisely the boundary between successful propagation and failure of the detonation wave [20,23–27].

In the past, a number of criteria have been proposed for which could determine the detonation limits, see Refs. [18,20,28] for a comprehensive review. Among those is the $\lambda/3$ rule. As the limits are approached, it is observed experimentally that the cellular detonation tends toward the lowest propagation mode of single-headed spin with $\lambda = \pi \cdot D$, where λ denotes the detonation cell size and D is the tube diameter. Hence, using the onset of single-headed spin detonation as the criterion for defining the detonation limits results in the proposition of the $\lambda/3$ rule. Recently, this criterion is re-examined and found to be applicable for various hydrocarbon mixtures and over a wide range of tube diameters [19].

In the literature, relatively very few detailed study of detonation limits for hydrogen mixtures had been made. In this study, new experiments were performed to observe the detonation velocity deficit and limits for hydrogen mixtures in small tubes. Three small diameter round tubes were used to ensure propagation of detonation wave over long distances ranging from 275 to 1667 tube diameter. Detonation velocities were measured by ionization probes located along the small tube. Two types of mixtures including the “stable” ($2H_2 + O_2 + 3Ar$) and “unstable” ($2H_2 + O_2$) mixtures were tested. For $2H_2+O_2+3Ar$ mixture, the detonation wave generally has a regular cellular structure; whereas for $2H_2+O_2$ mixture, the detonation front is characterized by an irregular cellular pattern [29]. The experimentally measured velocity deficit is also compared with the theoretical prediction using the Fay model, which is essentially based on the loss due to the flow divergence caused by the effect of negative displacement thickness of the wall boundary layer. Using the present results, this study further establishes the applicability of the onset of single-headed spin detonation or the $\lambda/3$ rule as a criterion to be defined for detonation limits for hydrogen mixtures [19,23,24,30–33].

Experimental details

The experimental apparatus consists of a driver section tube attached to the smaller diameter test section tube, as shown in Fig. 1. The driver section has a 25.4-mm inner diameter and 1500-mm length. Experiments were performed for the test section tubes of the same length $L = 3000$ mm but for different diameters: $L/D = 1667$, 652 and 275 for $D = 1.8$, 4.6 and 10.9 mm and the tube wall thickness $T = 0.7$ mm, 0.9 mm, 0.9 mm, respectively. Each test section tube protruded some distance into the driver section in order to mitigate the effect of wave reflection from the flange at the driver end (see Fig. 1). A spark plug was connected to the ignition system in the driver section to generate a high energy spark as in our previous studies [34–36,50]. A short length of Shchelkin spiral was also inserted just downstream of the spark plug to promote detonation formation.

Two combustible mixtures of $2H_2+O_2+3Ar$ and $2H_2 + O_2$ were tested in the present study. The argon-diluted mixture gives a stable detonation with regular cellular pattern while the undiluted mixture is usually characterized by an irregular cellular pattern [29]. The explosive mixtures were prepared via partial pressure method in a separated gas bottle, and were allowed to mix by diffusion for at least 24 h to ensure the homogeneity prior to be used. An Omega model PX309-030AI pressure transducer (0–206 kPa) with an accuracy of ±0.25% full scale was used to monitor the initial pressure in the detonation tube. The digital meter was calibrated to display