

Explosion Limits of Hydrogen-Oxygen-Steam Mixtures at Elevated Pressures and Temperatures

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1 Introduction

The radiolysis gas consisting of a stoichiometric hydrogen-oxygen mixture diluted with steam at different temperatures and pressures is a typical gas composition for the piping system of Boiling Water Reactors (BWR). There are significant gaps in the experimental data on radiolysis gas explosions relevant to BWR operating conditions: initial pressure up to 70 bar and temperature up to 570 K. No experimental data are available on the flame properties, critical conditions for the flame acceleration and detonation onset of radiolysis gas-steam mixtures at elevated pressures and temperatures. There are only known experimental data on flammability limits directly obtained for radiolysis gas-steam mixtures in the temperature range of 383-573 K and pressure range of 1-72 bar [1, 2]. Laminar burning velocities for stoichiometric hydrogen-oxygen mixtures diluted with steam were experimentally evaluated using a burner method at normal pressure and at the temperature of 373 K [3] and recently, using a spherical bomb method, at elevated pressures up to 70 bar and temperatures up to 573 K [4].

Critical conditions for the flame acceleration and detonation onset of stoichiometric hydrogen-oxygen-steam mixtures at pressures up to 70 bar and temperatures up to 573 K have been calculated in [5] using so called σ -criterion for the flame acceleration [6] and 7λ -criterion for the DDT [7]. A problem was that critical conditions for the flame acceleration and DDT were only based on experimental data obtained at initial pressures up to 3 bar.

The goal of this paper is to obtain new experimental data on critical conditions for the effective flame acceleration and deflagration-to-detonation transitions for hydrogen-oxygen-steam mixtures at elevated initial pressures up to 70 bar and temperatures up to 573 K in order to extend the working range of the models used the σ - and 7λ -criteria. Results of the work are required for BWR safety application and numerical codes validation.

2 Calculations

In terms of hazard potential and according to [6], three characteristic flame propagation regimes can be distinguished in obstructed channels: (1) "slow" or subsonic flames; (2) "fast" or sonic flames; (3) detonations. A state diagram of flame propagation regimes, based on the experimental data analysis and using the σ -criterion

for flame acceleration [6] and the 7λ -criterion for DDT [7], has been calculated for a triple hydrogen-oxygen-steam mixture at different pressures and temperatures. Fig. 1 shows calculated flammability limits (FL), flame acceleration (FA) limits and DDT limits at the characteristic scale $L=100$ mm for hydrogen-oxygen mixtures at the initial temperature of 300C and for two different initial pressures. Dotted line corresponds to the stoichiometric hydrogen-oxygen mixture ($\phi = 1$) diluted with steam (same as radiolysis gas (RG)). Mixtures outside the flammability limits are inert. As it seen from Fig. 1, the DDT domain at elevated pressure of 70 bar becomes practically the same size as the flame acceleration domain. That is, due to the promoting effect of initial pressure on the detonation onset, practically all the accelerated to sonic velocity mixtures can detonate at 70 bar of initial pressure and the DDT limits will be practically controlled by the flame acceleration limits.

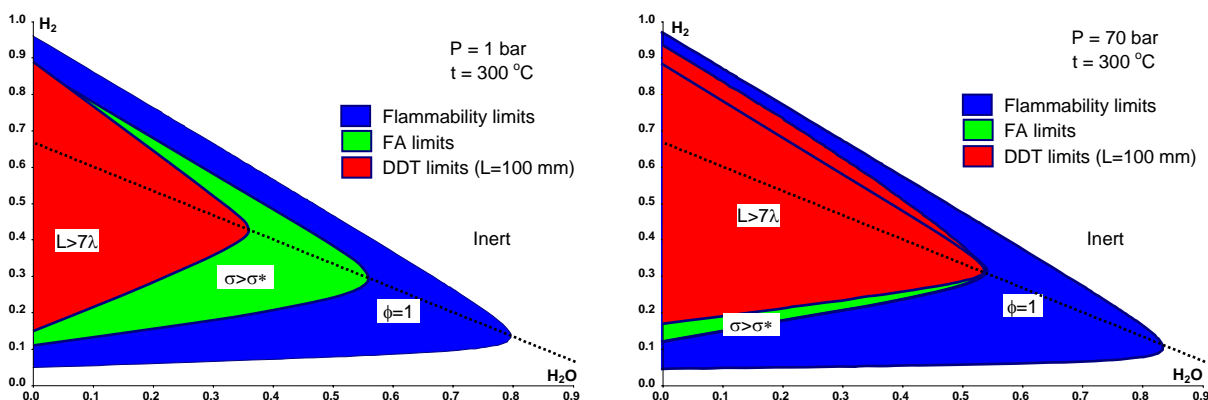


Fig. 1. Explosion limits diagram for hydrogen-oxygen-steam mixtures at 300C: 1 bar (left); 70 bar (right).

For such a diagram (Fig. 1) we can distinguish three specific points to characterize one of the flame propagation regime zones: (a) lower hydrogen concentration $[H_2]$ for lean mixtures ($\phi < 1$); (b) upper hydrogen concentration $[H_2]$ for rich mixtures ($\phi > 1$); upper steam concentration for a stoichiometric mixture ($\phi = 1$). Table 1 contains data on calculated flame propagation limits at $t = 300$ C and $p = 70$ bar for hydrogen-oxygen-steam mixtures. DDT limits are given for the characteristic size $L = 100$ mm. For the case when FA limit is narrower than DDT limit, the DDT limit has to be inside the FA domain. In the Table 1 expected DDT limits are given in red color.

Table 1: Flame propagation regime limits for hydrogen-oxygen mixtures ($p = 70$ bar, $t = 300$ C)

Limits	FL	FA	DDT
Lower $[H_2]$ ($\phi < 1$)	4.5%	10%	17-18%
Upper $[H_2]$ ($\phi > 1$)	96%	87-88%	92-93%
Upper $[H_2O]$ ($\phi = 1$)	84%	54-56%	53-54%

3 Experimental details

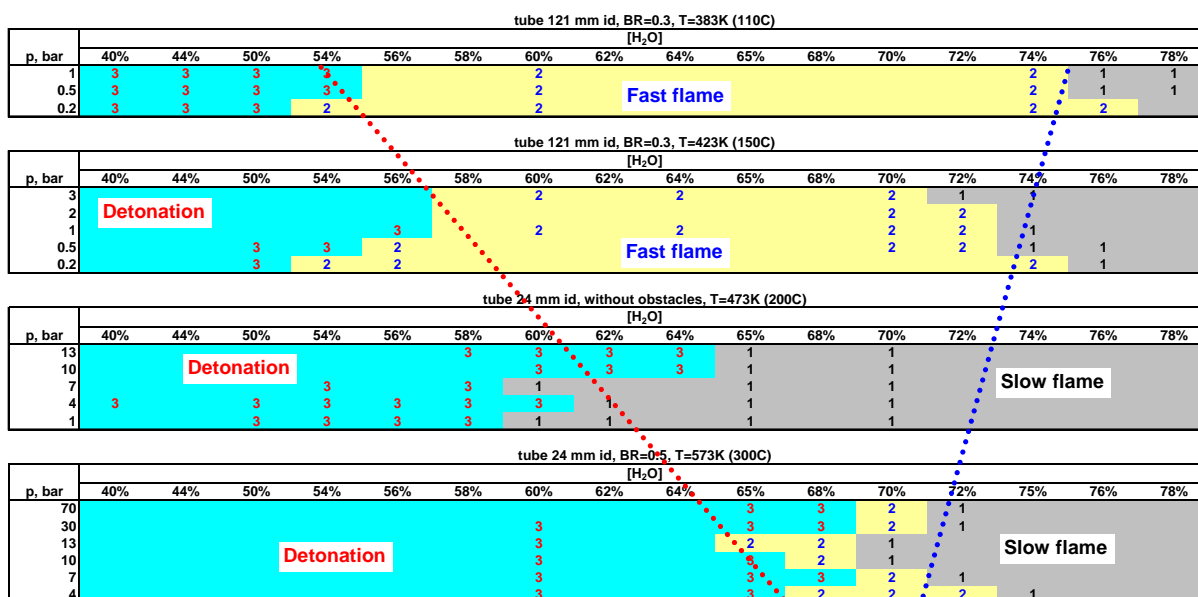
The experiments on flame acceleration and DDT limits as a function of pressure and temperature have been performed with hydrogen-oxygen-steam mixtures at pressures up to 70 bar and temperatures 110C, 150C, 200C and 300C. Two heated stainless tubes were used for this purpose: (1) 121 mm i.d. 8-m long tube; (2) 24 mm i.d. 4-m long tube. Ring shaped obstacles with blockage ratio $BR = 0.3$ or Schelkin spiral with $BR = 0.5$ spaced by tube diameter were installed in the tests to promote an effective flame acceleration. Test mixtures up to 3 bar of initial pressure were prepared by precise partial pressure method. The mixing time was about 8-12 hours.

Hydrogen-oxygen-steam mixtures with initial pressures in the range of 4-70 bar were prepared *in situ* using mass flow rate controllers for water, hydrogen and oxygen. Main problems for the mixture preparing were to avoid steam condensation and to take into account steam nonideality at elevated pressures. The heating system of the tube supported longitudinal temperature uniformity with an accuracy ± 2 deg. The ignition of test mixtures was provided by means spark or glow plugs. The spark energy was changed from 200 mJ to 180 J depending on the sensitivity of the mixture to be ignited. Light signal from the flame front and pressure signal were recorded by photodiodes and pressure sensors (KISTLER design) installed along the tube.

4 Data analysis

Three typical flame propagation regimes have been occurred in the experiments: (1) slow subsonic flames; (2) fast sonic or “choked” flames; (3) detonations or “quasi-detonations”. Main experimental results on flame propagation regimes as function of initial pressure and temperature are summarized in Table 2. As it follows from Table 2, the pressure and temperature increase has significant promoting effect on critical detonation conditions because the detonation cell size of hydrogen-oxygen-steam mixtures reduces with a pressure and temperature increase. Critical detonation conditions for initial temperatures 473K and 573K would be shifted to the less reactive mixture (with higher steam dilutions) because 5 times smaller tube was used in these tests compared to that for temperatures 383K and 423K. Flame acceleration limits have an opposite trend with pressure and temperature increase. High initial pressure and temperature suppresses flame acceleration. Compared to the DDT-limit, it results in opposite trend of the border line between slow and fast flames as function of pressure and temperature.

Table 2: Flame propagation regime limits for hydrogen-oxygen mixtures



Qualitatively, experimental critical conditions for the flame acceleration and for the detonation onset have the same trend against temperature and pressure changes compared to our preliminary calculations but quantitatively the difference of predicted and experimental limits is very significant. For instance, our preliminary calculations for large tube (121 mm i.d.) give a DDT limit of 47% H₂O and flame acceleration limit of 72% H₂O at 3 bar and 423K. The experimental flame acceleration limit is consistent with calculations but the experimental DDT limit of 56-60 % H₂O is much higher than calculations. The difference between calculated and experimental DDT limits for the small tube (24 mm i.d.) at $p_0 = 70$ bar and $T_0 = 573$ K is much larger: 48% and 68% H₂O

correspondingly. The reason is that the detonation cell width $\lambda = 10$ cm calculated for hydrogen-oxygen-steam mixture with 68% H_2O at $p_0 = 70$ bar and $T_0 = 573\text{K}$ is more than 10 times larger than value of $\lambda = 8$ mm estimated from the experiments.

6 Conclusions

Experiments on flame acceleration and detonability limits for hydrogen-oxygen-steam mixtures in wide range of initial pressures (1-70 bar) and temperatures (383-573K) have been performed. It was shown that initial pressure and temperature have significant promoting effect on critical conditions for detonation onset. The flame acceleration limit has an opposite dependence vs. initial pressure and temperature. It leads to shrink of the gap between flame acceleration and detonability limits. As a result the detonation onset at elevated pressures and temperatures could be controlled by the conditions for flame acceleration.

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