Hydrogen explosion suppression in experiments of different scale

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1 Introduction

There are many ways to suppress hydrogen explosion in a closed volume: creation of inert atmosphere, usage of different types of recombiners, injection of phlegmatizator additives etc. Phlegmatization effectiveness of different compounds and mixtures are not widely tested at different scales and initial conditions.

Available systems that remove hydrogen from enclosure have limited performance. Formation of explosive hydrogen air mixtures with steam is possible in emergency situation for some rooms inside NPP containment. Taking into account that maximum operational pressure could be up to 4.8 atm even the slowest combustion mode – laminar burning can be extremely dangerous. Thus in our work the attempt of full hydrogen-air mixture combustion suppression with special phlegmatizing additives was done. Effect of scale for this problem is essential especially in a question of ignition source. One should be sure that energy of source is high enough to ignite mixture. If ignition source is small it is possible that combustion decays because of high heat losses and curvature of flame front during initial stage of combustion process. This is one of the main reasons that results of small scale experiments should be confirmed in a medium and large scale experiments.

Preliminary experiments on hydrogen air mixture phlegmatization were performed in a small scale facility to minimize expenses and to test wider range of phlegmatizatior substances. Binary and triple mixtures of perspective individual substances were combined to obtain better phlegmatization effect in a last series of experiments. Best individual substances and binary mixtures were selected for following medium and large scale test.

2 Experimental

Small scale experiments were conducted in a heated tube of 1 m length, 66 mm internal diameter, total volume 3.5 l. Initial temperature in experiments varied from 20 to 120˚C, initial pressure was 1 atm. Four pressure gauges and 10 light sensors were installed in facility. Ignition was performed with heated platinum wire of 15 mm length and 0.8 mm diameter, estimated temperature of the wire *T*~1500˚C.

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Figure 1. Small scale experimental facility.

Medium scale facility is a heated tube of 8 m length, internal diameter 121 mm, total volume 92 l. Initial temperature in experiments varied from 20 to 120˚C, initial pressure was 1 atm. Ignition was performed with heated nichrome surface 30 cm^2, estimated temperature T~1500˚C. Registration system of medium scale facility consists of 16 collimated photodiodes and 1 tensoresistive pressure gauge.

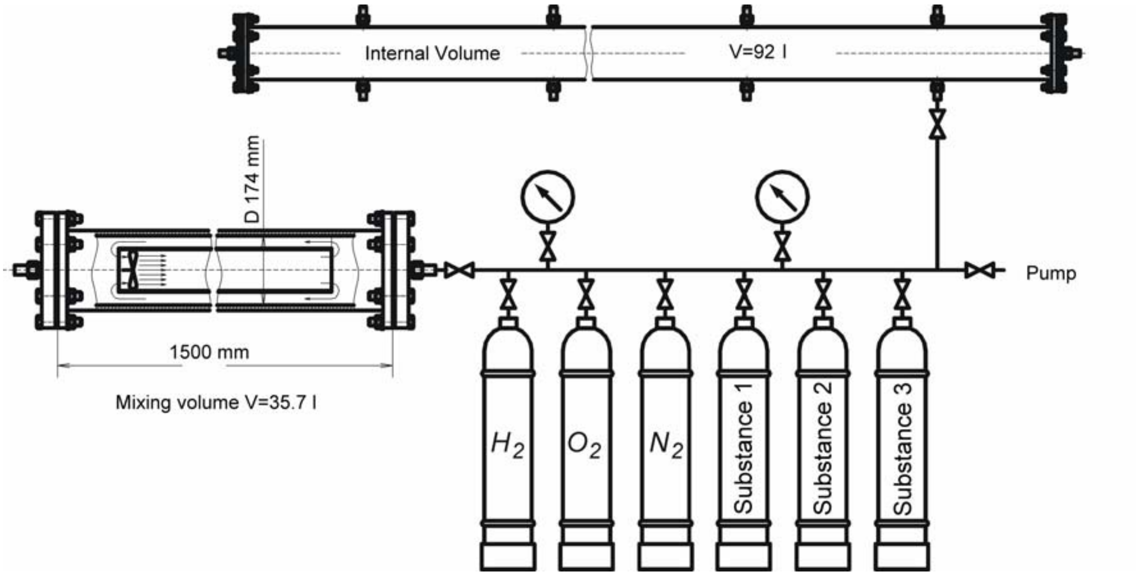

Figure 2. Scheme of medium experimental facility.

Total volume of large scale experimental facility is 10.6 m^3. Working volume of experimental facility is semicylinder 1.5 m internal diameter covered with 120 mkm polyethylene film. Ignition system consists of nichrome plates, total square area 200 cm^2, heating time <100ms, estimated temperature T~1200˚C. Initial temperature in experiments was 20˚C, initial pressure - 1 atm. Data acquisition system recorded the following data in the experiments: high-speed video recording of combustion processes and combustion and explosion parameters (pressure and light data).

Figure 3. Overall view of large scale facility

3 Results and Discussion

Mechanism of combustion inhibition by halogenides is based on suppression of chain process and active radicals fastening. Eleven individual substances were selected as perspective phlegmatizators for small scale tests. Stoichiometric hydrogen-air mixture was used as a combustible mixture in all experiments. Ability to suppress stoichiometric hydrogen-air mixture combustion with less than 20% addition of phlegmatizator was treated as criteria of substance effectiveness. Small addition of substance that appreciably reduce combustion velocity of hydrogen-air mixture, but not capable to suppress combustion was treated as perspective additional component.
11 individual substances, 5 binary mixtures and 1 triple mixture were tested in a small scale experiments. One individual substance ($C_2F_4Br_2$) and two binary mixtures ($C_2F_4Br_2$ + $C_2F_3Cl_3$ and $C_2F_4Br_2$ + CF_3I) were selected for medium and large scale tests.

Individual substance $C_2F_4Br_2$

Results of hydrogen-air mixture phlegmatization by $C_2F_4Br_2$ in a small scale tests are presented in Table 1.

Table 1. Experimental results with addition of $C_2F_4Br_2$ in a small scale tests.

Phlegmatizator concentration, %	Initial temperature, ^oC	Maximum overpressure, bar	Flame velocity, m/s	combustion
3	23	5.5	31	burn
10	23	2.3	1.5	burn
11	**23**	**0.4**	**0**	**decay**
12	22	0.4	0	decay
13	22	0.2	0	decay

Flame velocity corresponds to average flame front velocity near the tube end. Full stoichiometric hydrogen-air mixture combustion suppression achieved at 11% addition of $C_2F_4Br_2$ for T=23°C in a small scale tests. For 14% addition of $C_2F_4Br_2$ there is no light signal even on a first light sensor located at 5cm from ignition point. Full stoichiometric hydrogen-air mixture combustion suppression achieved at 11% addition of $C_2F_4Br_2$ for T=120°C.

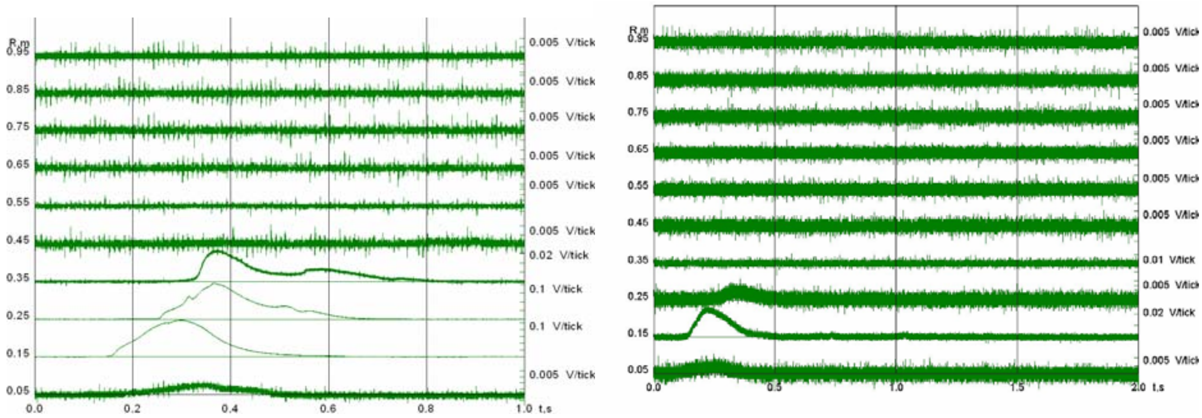

Figure 4. X-t diagram of combustion decay for 11% addition of $C_2F_4Br_2$ and T=23°C (left) and for 14% addition of $C_2F_4Br_2$ and T=120°C (right) in a small scale tests.

Medium scale experiments confirmed minimum $C_2F_4Br_2$ concentration values that allow stoichiometric hydrogen-air mixture full combustion suppression for initial temperature T=23°C and T=120°C.

Individual substance CF_3l

Results of hydrogen-air mixture phlegmatization by CF_3l in a small scale tests are presented in Table 2.

Table 2. Experimental results CF_3l with addition of in a small scale tests.

CF_3l, %	Initial temperature, ^oC	Maximum overpressure, bar	Flame velocity, m/s	combustion
10	25	2	1	Burn
15	25	2.2	0.9	Burn
20	25	1.2	0.8	Burn

Flame velocity – average flame front velocity near the tube end. Full stoichiometric hydrogen-air mixture combustion suppression did not achieve in a small scale tests with addition of CF_3l for T=25°C. But effective flame velocity reduction was observed. Because of chemical properties CF_3l can be treated as perspective additional component in phlegmatization binary mixture.

Binary mixture $C_2F_4Br_2$ + CF_3l

Results of hydrogen-air mixture phlegmatization by binary mixture $C_2F_4Br_2$ + CF_3l in a small scale tests are presented in Table 3.

Table 3. Experimental results for binary phlegmatizator mixture $C_2F_4Br_2$ + CF_3l in a small scale tests.

$C_2F_4Br_2$ + CF_3Cl, %	Total concentration,%	Initial temperature, ^oC	combustion
8% + 2%	10	27	burn
9% + 1%	10	24	burn
7% + 3%	10	26	burn
9% + 2%	**11**	**27**	**decay**

Experiments with binary mixtures showed that binary mixture has the same phlegmatizing concentration as individual substance $C_2F_4Br_2$ for 20°C and lower phlegmatizing concentration than $C_2F_4Br_2$ for 120°C.

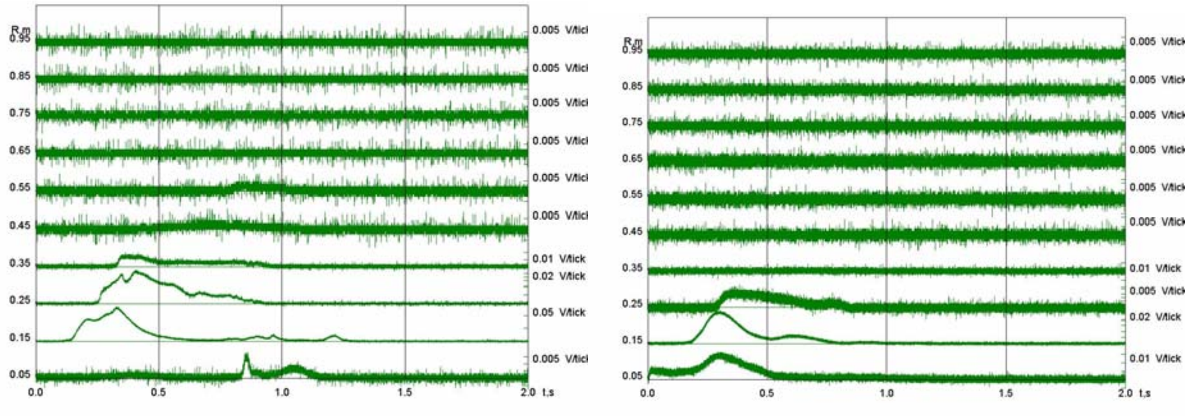

Figure 5. X-t diagram of combustion decay for 9% $C_2F_4Br_2$ + 2% CF_3l and T=27°C (left) and for 10% $C_2F_4Br_2$ + 3% CF_3l and T=120°C (right).

Medium scale experiments confirmed possibility of stoichiometric hydrogen-air mixture full combustion suppression for initial temperature T=27°C and T=120°C for binary mixture $C_2F_4Br_2$ + CF_3l with higher phlegmatizator concentrations than in a small scale experiments.

Large scale experiments confirmed possibility of stoichiometric hydrogen-air mixture full combustion suppression for individual $C_2F_4Br_2$ and for binary mixture $C_2F_4Br_2$ + CF_3l, but with higher phlegmatizator concentrations than in a small scale and medium scale experiments.

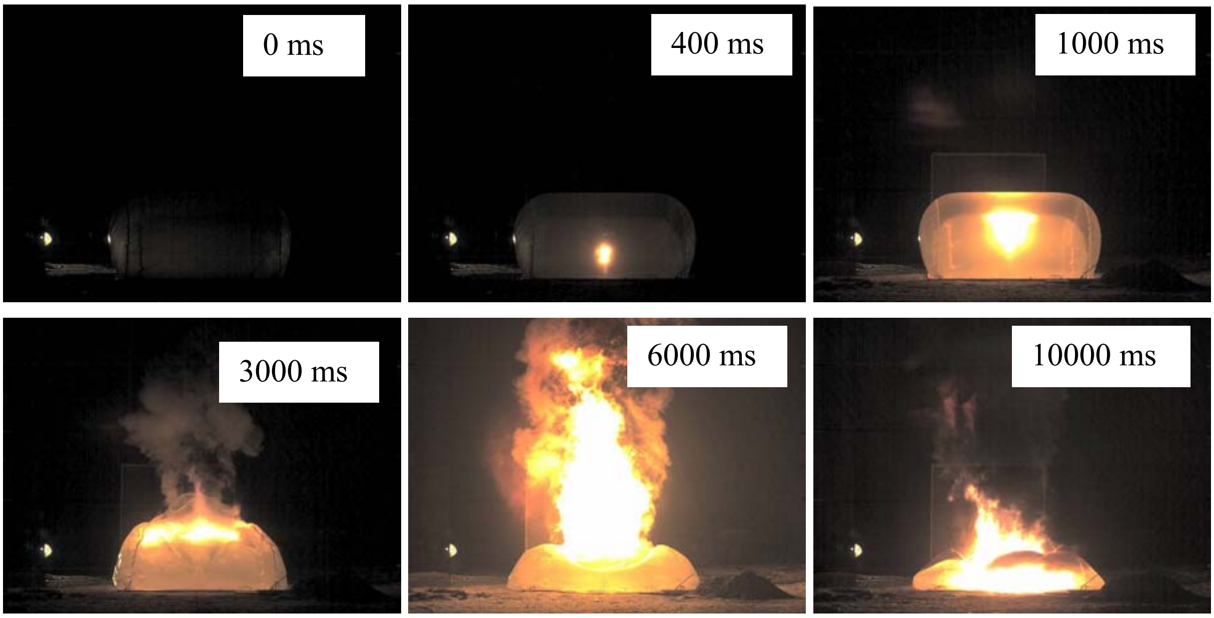

Figure 6. Combustion of stoichiometric hydrogen-air mixture with addition of 8.5% $C_2F_4Br_2$ phlegmatizator. High-speed video record frames.

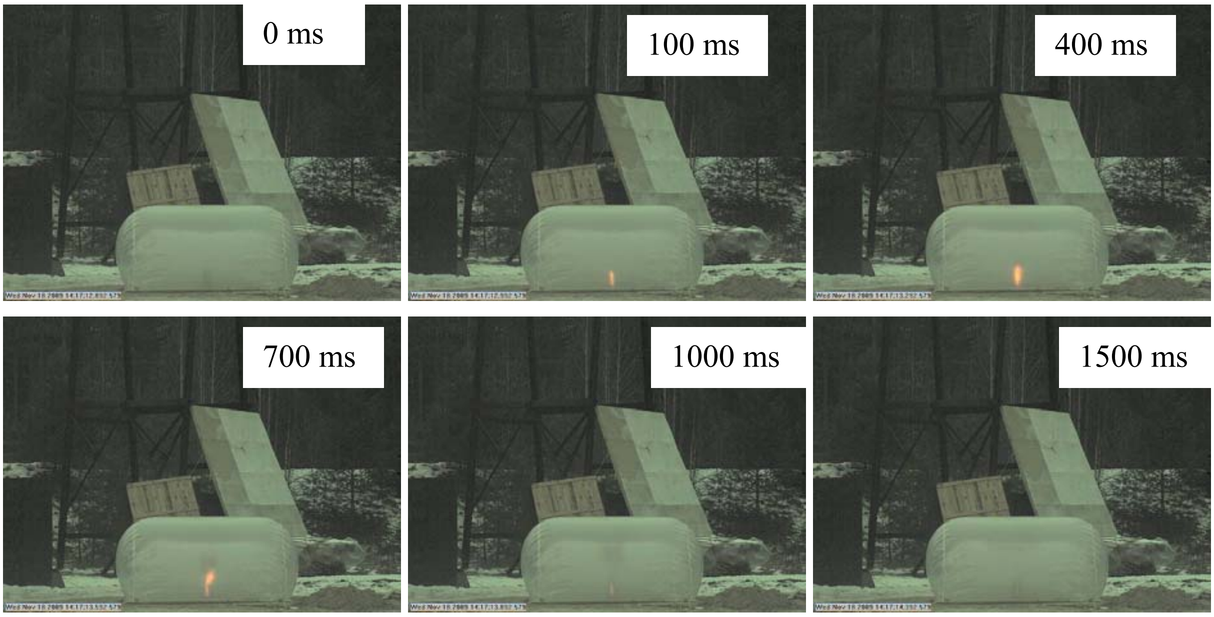

Figure 7. Full combustion suppression of stoichiometric hydrogen-air mixture with addition of 10.4% $C_2F_4Br_2$ + 1.7% $C_2F_3Cl_3$ phlegmatizator mixture. High-speed video record frames.

4 Conclusions

Hydrogen mixture phlegmatization experiments of different scale showed that:

- Eleven individual compounds, five binary mixtures and one triple mixture were tested as phlegmatization additives
- Binary mixture that fully suppresses hydrogen combustion was found. Its minimal concentration was 12% at T=20°C and 14% at T=120°C
- Effectiveness of the mixture to suppress hydrogen combustion was confirmed in experiments of three different scale
- Effectiveness of the mixture in wide temperature and pressure range and different geometrical conditions needs further investigation.

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