Hydrogen Combustion in the Course of a Loss-of-Coolant Accident in Nuclear Power Plants

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Introduction

In the course of a hypothetical severe accident in a light water reactor large amounts of hydrogen can be released caused by oxidation of the zirconium inventory with the water present and by core-concrete interaction, following a failure of the pressure vessel. This hydrogen suffices to form a combustible mixture together with the air and vapour in the containment. Carbonmonoxide and carbondioxide are also present in rather high quantities. Investigations as to whether the integrity of the containment risks being endangered by combustion of this hydrogen presently constitute an important sector in reactor safety.

Combustion Modes

The presence of adequate sources to ignite a possibly arising combustible mixture sooner or later can be assumed, as combustible hydrogen mixtures are characterized by very low ignition energies; the required ignition temperatures, however, are comparably high. The following basic reaction and combustion modes can be distinguished:

Quiescent Oxidation

In the case of a high mixture temperature (about 500°C), lacking an ignition source, slow oxidation of the hydrogen can take place without a significant flame.

Diffusion Flame

The formed hydrogen burns off continuously in a locally more or less stable diffusion flame, provided an ignition source near the location of the discharged hydrogen and sufficient oxygen supply are present. Such a flame does not produce significant pressure waves. The containment structures act as heat sinks, thus rendering the pressure increase due to the released energy and the consequent temperature rise in the containment atmosphere to be low.

Combustion under Pre-mixed Conditions

Without an ignition source or sufficient oxygen supply near the discharge location, the formed hydrogen disperses in the containment atmosphere. Ignition leads to a combustion under pre-mixed conditions, which is a very rapid process. Basicly two combustion mechanisms can be distinguished under premixed conditiones:

- Deflagration
  In the case of a deflagration the flame propagates by heat and mass transfer mechanisms at the flame front, i.e. the not yet burned mixture in front of the flame is heated up by heat transfer and diffusion to reaction temperature before it reacts itself. The flame front velocity is a function of the flow condition. In laminar flow it amounts to few m/s. Turbulence can lead to a decisive acceleration up to several hundred m/s, due to an increase of the flame surface. A pressure wave is formed ahead the flame front. In very rapid deflagrations, this pressure wave can reach a 10-fold excess when compared to initial pressure.
- Detonation
  In contrast to a deflagration the unburned mixture in a detonation is heated up by a strong shock wave, behind which the mixture reaches self-ignition temperature and reacts after a certain induction time. Shock wave and reaction zone are strongly interdependent and propagate into the unburned mixture with multiple speed of sound. The velocity of a detonation varies depending on the mixture composition. In hydrogen-air-mixtures, for example, it reaches 2 km/s.

Deflagration-Detonation Transition (DDT)

For deflagrative ignition of the observed mixtures very low energies suffice, as already mentioned above. On the other hand, very powerful ignition sources are needed for direct initiation of a detonation, which release great quantities of energy in a very short time. In practice this is only possible with explosives. Such powerful sources of ignition are not to be expected in the containment rendering only deflagrative ignition relevant for safety considerations. Basically, an at first slowly propagating deflagration can under the influence of turbulence inducing obstacles strongly accelerate, and even, under certain circumstances, undergo a transition to a detonation. Due to the resulting strong shock wave, these processes constitute the main focus of reactor safety. Turbulence inducing obstacles in the case of a combustion can be, for example, installations in rooms and especially openings between two rooms, i.e. doors or flaps.

Qualitatively, the fundamental processes of turbulent flame acceleration are known: starting at the ignition source, the flame front propagates into the combustible mixture. The hot gas clouds behind the flame front cause an expansion flow which leads to a generation of turbulence by obstacles in the flow. This has the effect of a folding of the reaction zone which is equivalent to an enlargement of the flame surface. As a consequence, heat transfer from the reaction zone to the unburned mixture in front of it is enhanced and, thus, the flame is accelerated. This, in turn, results in a boost of the expansion flow and consequently a further increase of turbulence together with additional flame acceleration. This positive feedback cycle between turbulence initiation and flame acceleration can lead to a DDT, if quenching effects caused by too intensive mixing do not limit acceleration. In the first instance, the chances for a DDT depend on mixture composition. Experiments in a three-component mixture of hydrogen, air and vapour have yielded the limits shown in the ternary diagram of Fig. 1. Additionally the igniton limit is shown. Not all mixtures within the detonation limits are prone to detonation to the same extent. A DDT is rather unlikely to occur near the limits. The so-called detonation cell width denotes a characteristic value. Detonation cells develop by the process of minor shock waves overlapping the main impact wave which criss-cross the latter. The distance between these crossing shock waves is a function of mixture composition and the initial conditions. Their traces can be made visible on sooted foils which are arranged parallel to the direction of propagation. There the shock waves create a cellular pattern, the width of which is denoted as the detonation cell width λ. The smaller it is, the greater the mixture's detonation hazard. A stably propagating detonation requires a channel width of at least λ.

This geometrical influence is to be noted for laboratory experiments, for real geometries, however, it has hardly any limiting effect. Even if the mixture composition is favourable for a DDT, this alone does not necessarily mean that a DDT will actually occur. Geometry, moreover, plays a significant role in the turbulent flame acceleration preceding a DDT. Modeling the turbulent combustion is presently only possible for special, strictly defined application cases, due to problems understanding turbulent flows as such, without reaction, and the additional interaction of flow and reaction.

Research Work

Lacking the possibility of a quantitative theoretical forecast of turbulent combustion processes, mainly the results of experimental work have to be relied on.

Phenomenological Tests

Combustion experiments mainly with dry hydrogen-air mixtures have been carried out in various scales. In laboratory experiments at the Lehrstuhl A für Thermodynamik of the Technical University of Munich the influence of different geometries on the combustion behaviour of various hydrogen-air-mixtures has been observed.

The test set-up comprised a 6m long closed tube, with a diameter of 66 mm (Fig.2), which was filled with defined mixtures. Ignition was actuated by a spark plug at one end of the tube. By means of an optoelectronic recording system, a time-distance diagram of the flame propagation was recorded and from this the flame velocity was determined. For the experiments, various 3 m long obstacle rows have been installed in the tube beginning at the tube end harbouring the implanted spark plug. They consisted of orifice-like obstacles arranged in equidistance, one behind the other. Obstacle rows with blockage ratios of 0.3 and 0.7 (free diameter surface to total diameter surface of the tube) were employed with obstacle spacings between 35 and 500 mm and hydrogen concentrations of 10-28%. In addition, measurements were carried out with a single orifice, arranged 100 mm away from the spark plug, and as a reference in an obstacle-free tube. The initial state for all experiments was set to 80°C and 1 bar. Fig. 3 exemplarily shows the flame velocities achieved in the explosion tube.

Mixtures with a hydrogen component of less than 10% could not be ignited reliably. With low hydrogen concentrations of 10-13% very high flame velocities occur in the obstacle rows and also after a single orifice in comparison to the obstacle-free tube. Quenching effects prevent combustion being sustained for the whole tube length. Obstacle rows with short spacing between obstacles feature a quicker flame acceleration than those with greater spacing. The quenching effects, on the other hand, come into force more rapidly, too.

Mixtures of a higher hydrogen concentration sustain combustion throughout the tube length in any case. Detonations have been observed in mixtures with a hydrogen concentration above 20%. This value goes confirm with experiments of other authors which have shown that stable detonations in tubes can only then be sustained, if the dimension of the detonation cell width is smaller than the tube diameter. Basically, however, all mixtures, in which a critical flame velocity can be achieved, can be classified as hazardous to detonation. In the case of high blockage ratios, small spacing between obstacles do not allow acceleration above the critical flame velocity. Obstacles with a small blockage ratio set at short spacing, however, result in the most powerful combustion processes that have been observed. Fig.4 surveys the maximum flame velocities measured in the obstacle rows as a function of hydrogen concentration and obstacle distance. The orifices accordingly have the following effect on the combustion process in the tube: besides the accelerating effect by induced turbulence, very close obstacles of high blockage ratios obviously prevent the development of a shock wave in front of the flame, which is a precondition for a supercritical flame velocity. All mixtures already produce a high flame acceleration with a single orifice, even though it is installed at a very small distance of only 100 mm from the ignition source, meaning a surely only moderate jet stream formation, before the flame front passes.

The overall result of the tests: the ignition quality of hydrogen-air-mixtures with a hydrogen percentage of under ten is rather bad with the weak ignition source used. On the other hand, in mixtures with 10% hydrogen and more a very strong combustion process can occur by turbulence and jet promoting obstacles. The initiation distances for a strong shock wave with a possible transition into a detonation are very short, thus rendering the impeding effect of obstacle on the development of a shock wave in the real geometries of a containment not relevant. Comparisons with large-scale experiments of other institutes (Battelle, Sandia/U.S.A.) show that predictions on basic combustion behaviour derived from experiments with the small set-up used here are very well possible for real geometries.

Based on these experiences experiments are presently being carried through at the Lehrstuhl A für Thermodynamik of the Technical University of Munich in the scope of a BMFT (Federal Ministry for Research and Technology) supported research project investigating the influence of increased initial temperatures in the range up to 280°C and various vapour concentrations in the combustion gas atmosphere using the experimental set-up described above. These experiments aim at determining the range limits of mixtures for which a powerful shock wave or even a DDT is basically possible under the initial conditions to be expected in reality (mixture composition, pressure, temperature). These results are used for further investigations in other institutes which are to test the large-scale acceleration effects in geometries closer to reality.

Investigations of Fundamentals of DDT and Turbulent Flame Acceleration

Investigations of fundamentals are necessary to achieve a full understanding of the processes of turbulent flame acceleration. For this reason, a model for the DDT has been developed in the Lehrstuhl A für Thermodynamik. Present work concentrates on the turbulent flame acceleration, a pre-requisite necessary for a DDT. In the scope of several BMFT projects the decisive interactions between flow and combustion processes are being investigated.

As the, in the above mentioned explosion tube, very rapidly progressing flame is very difficult to reach with measuring instruments, a part of the investigations is performed on stationary flames in a tube burner. Here concentration profiles in the flame front are recorded by means of a Raman scattering system in order to determine the structure of a turbulent flame and the flame thickness as a function of concentration and combustion velocity. Fig. 5 shows the result of the first measurements at the flame front. The local hydrogen concentration is plotted against the flame length under variation of the flow velocity and the initial hydrogen concentration. As yet it is not possible to correlate the results with other investigations.

Another part of the experimental work is done using the explosion tube under unstationary conditions. The expansion flow caused by the hot gas clouds behind the flame is measured by means of a laser-doppler velocimeter. Thus, the connection between turbulence of the flow and combustion velocity is to be investigated. Fig. 6 shows the temporal proceeding of the flow in the middle of the explosion tube in a combustion experiment. The highly turbulent flow in front of the flame is to be recognized and the flame passage through the measuring volume as a sudden change of velocity, due to the density jump in the reaction zone.

Final Remark

Scientists are still far from completely understanding turbulent combustion processes required for safety analysis, making further investigations necessary in this field. To guarantee highest possible safety based on the current knowledge, mitigation concepts are being discussed employing deliberate ignition before critical hydrogen concentrations are reached. These mixture ranges are known.

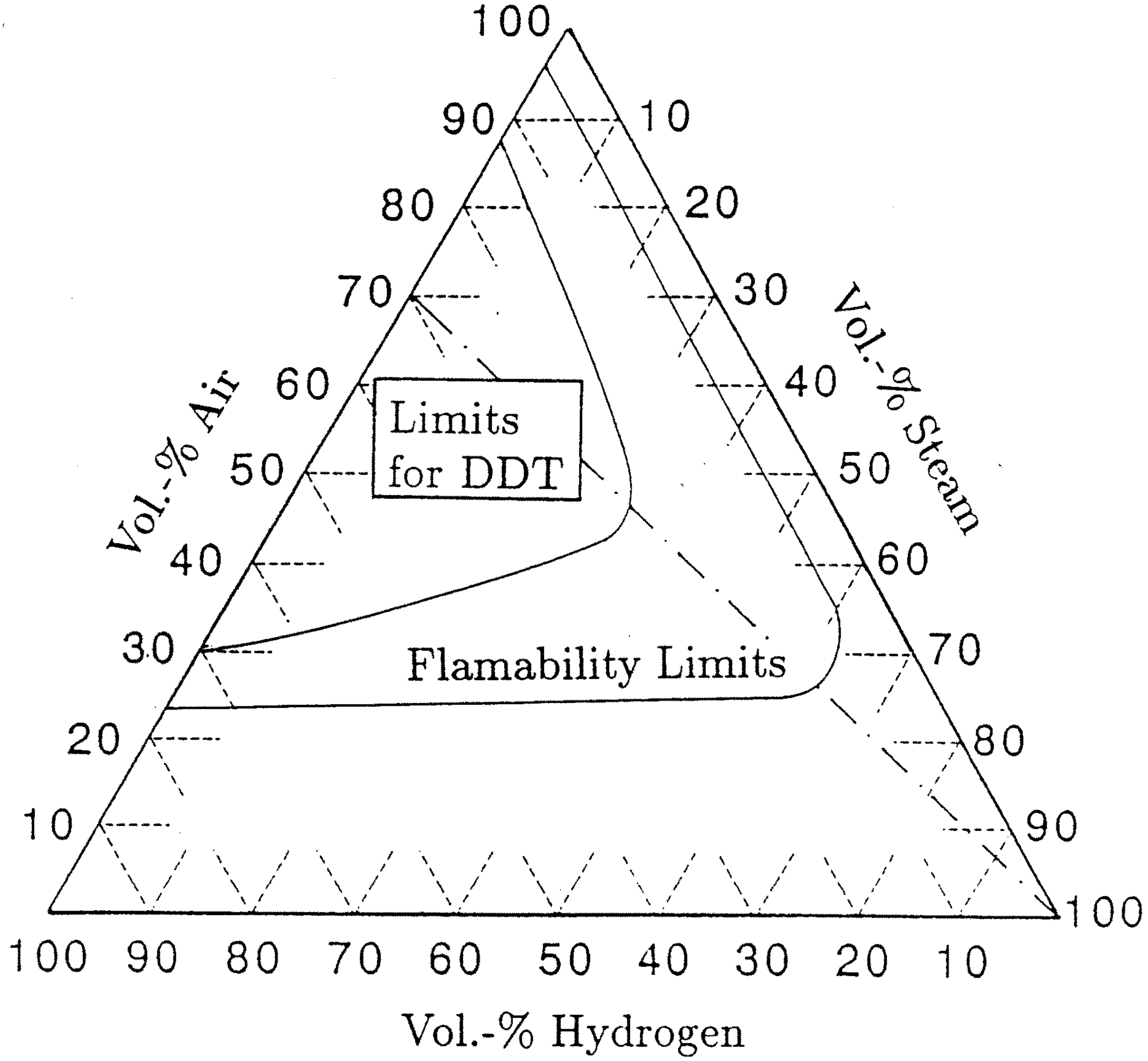

Fig. 1: Flamability limits and DDT limits in hydrogen-air-steam mixtures

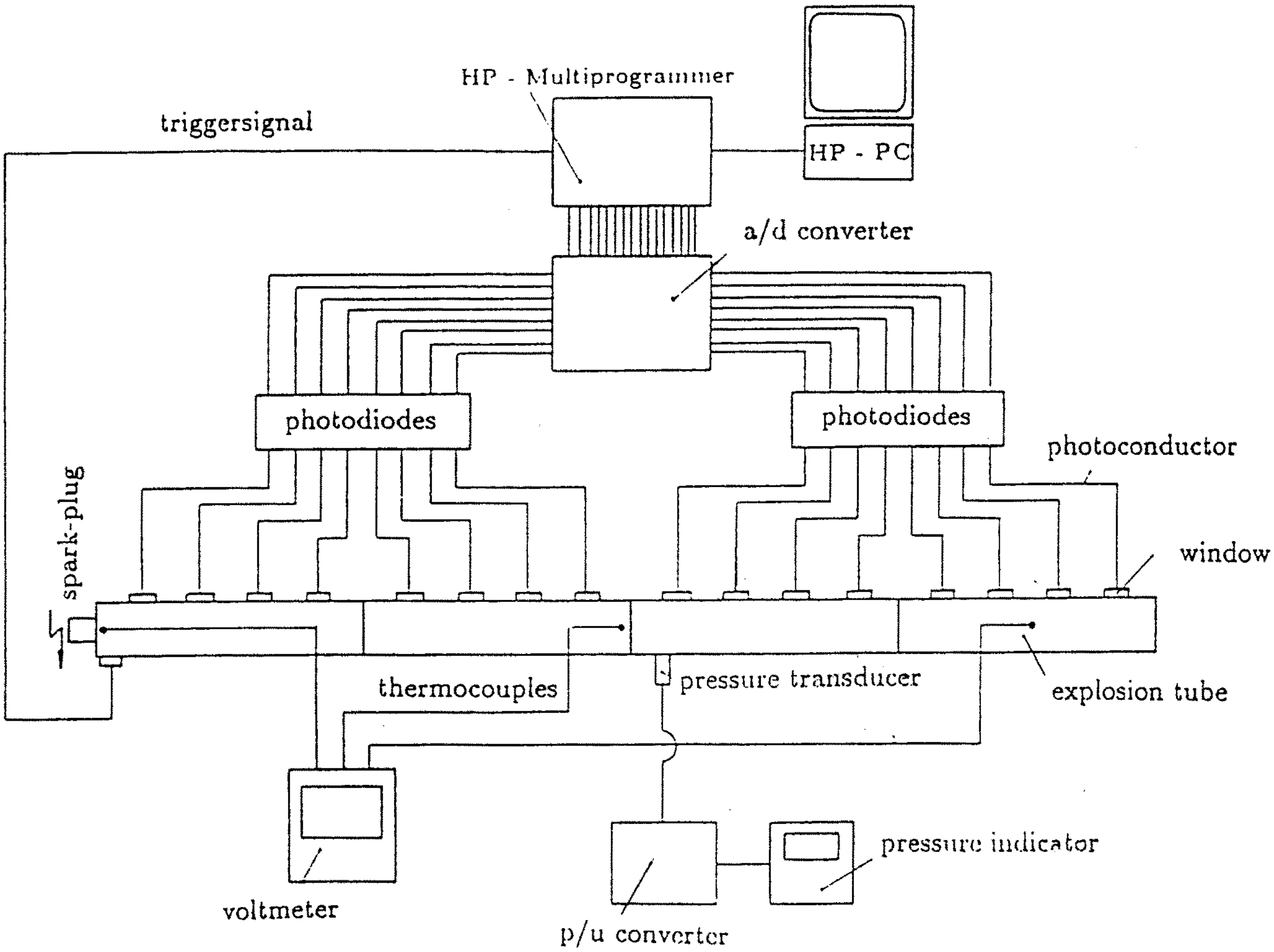

Fig. 2: Experimental setup with explosion tube and measuring arrangement

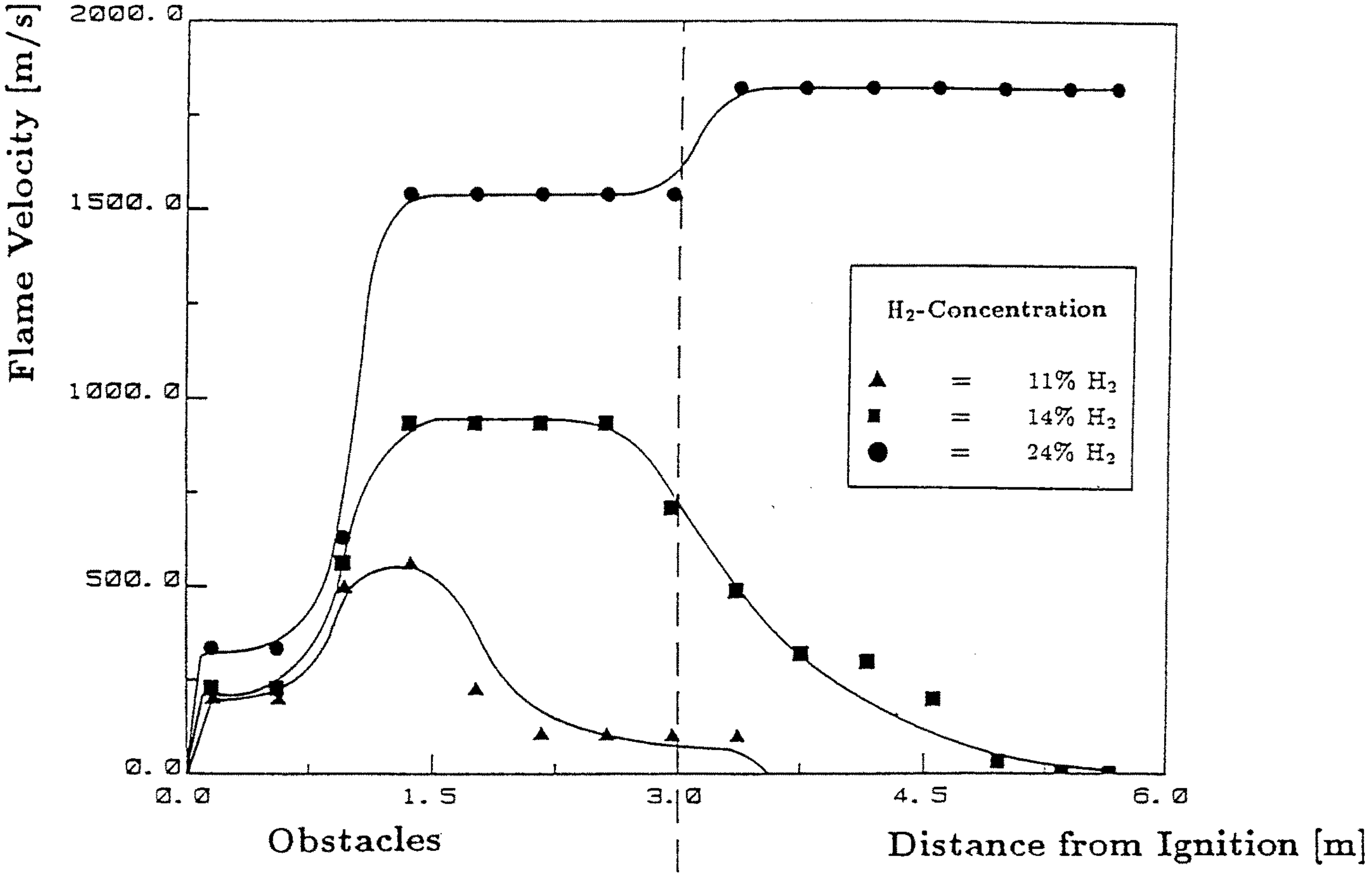

Fig. 3: Flame velocity in the explosion tube, obstacle spacing 490 mm, blockage ratio 0.7

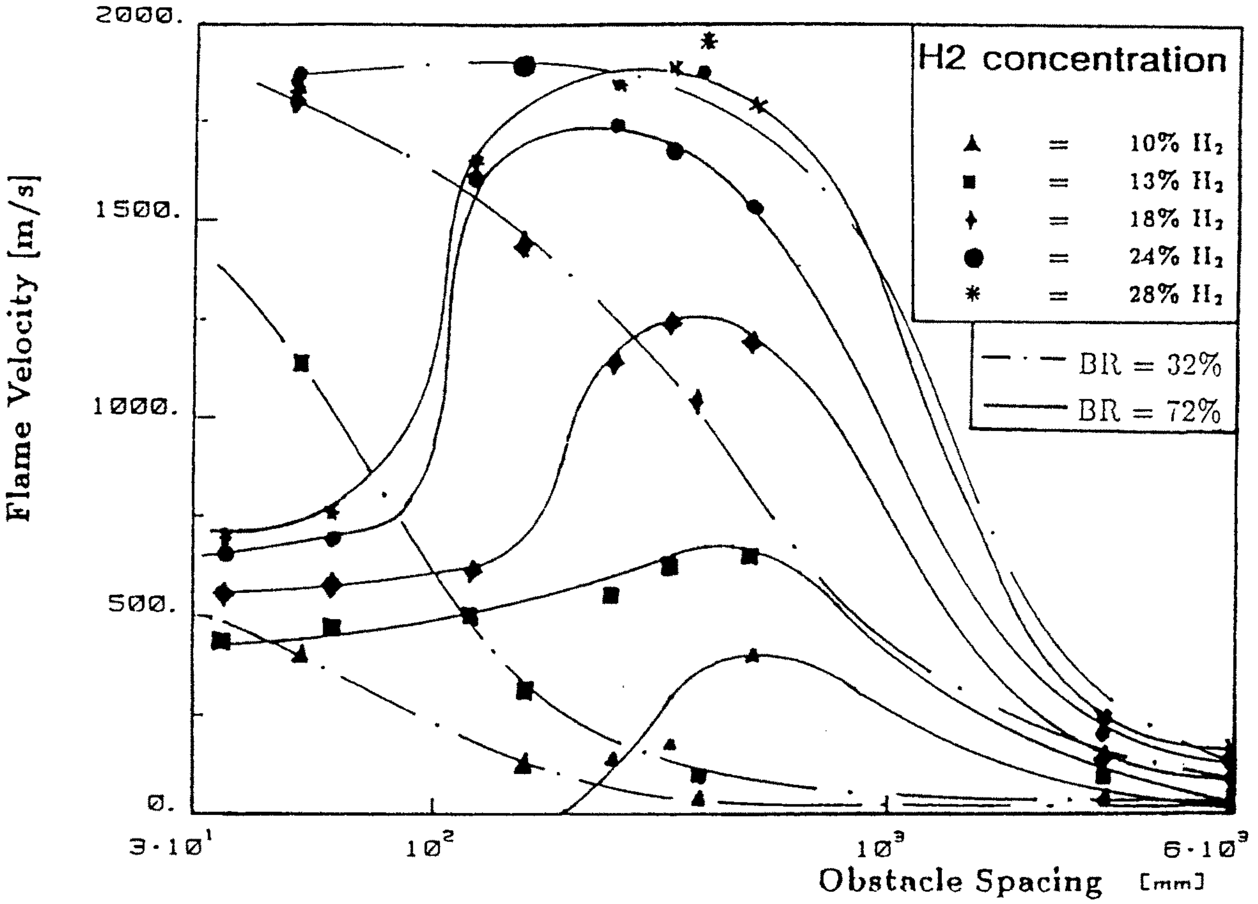

Fig. 4: Maximum flame velocity in the explosion tube as a function of hydrogen concentration, obstacle spacing, and blockage ratio

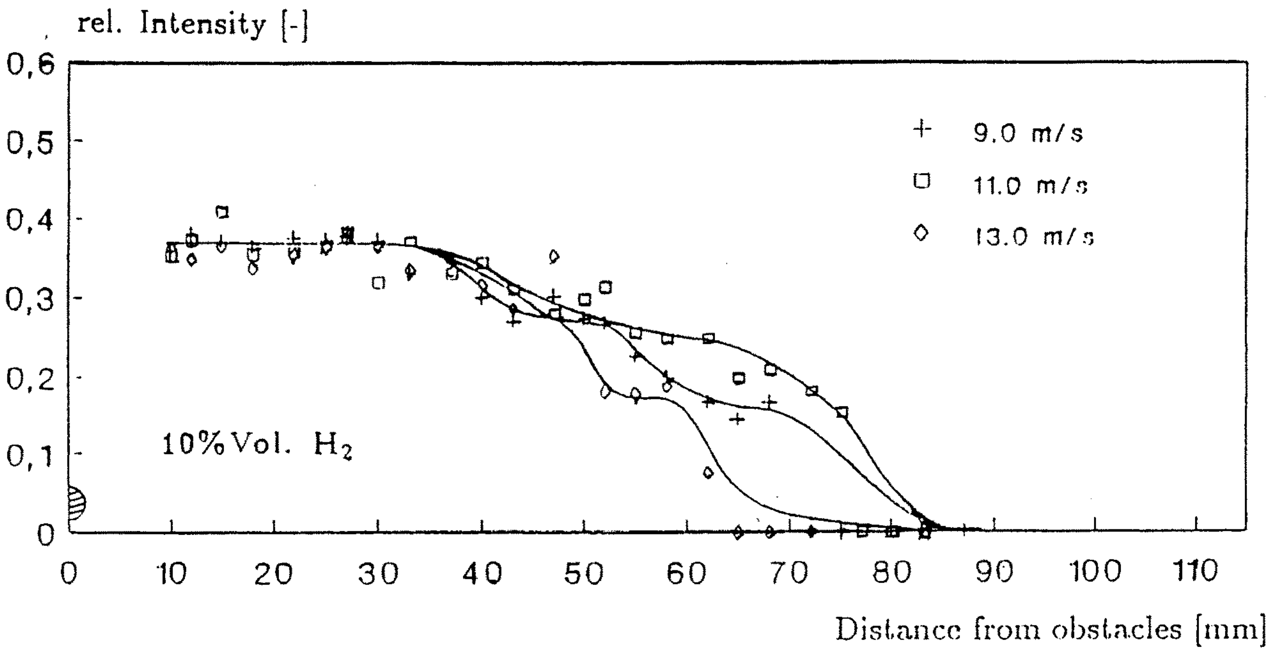

Fig. 5: Intensity of the H_2 raman signal (i.e. H_2 concentration) in a stationary premixed H_2-air flame downstream of turbulence inducing obstacles at initial gas velocities of 9, 11 and 13 m/s

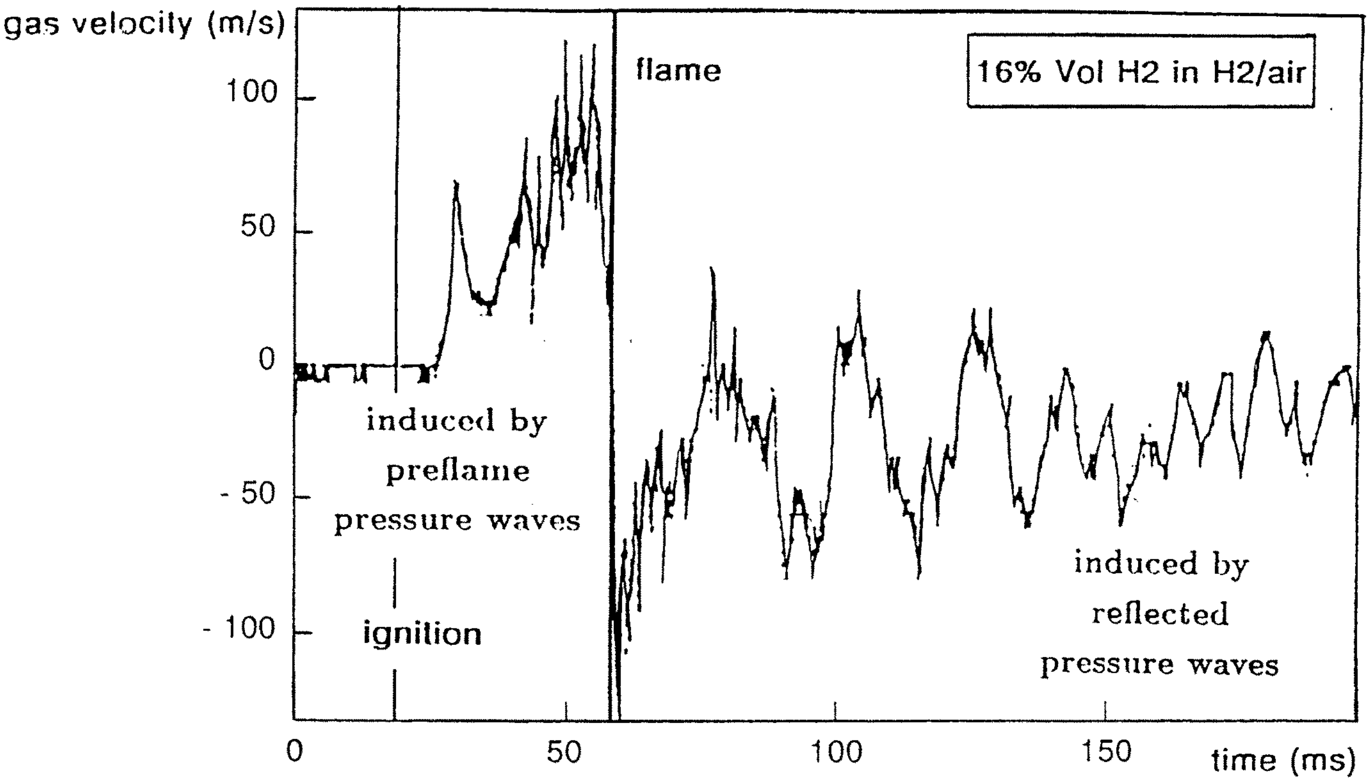

Fig. 6: local gas velocity recorded in the middle of the explosion tube during a run with a mixture of 16% H_2 in H_2-air