

SEVERE ACCIDENT IN A LIGHT WATER REACTOR: INFLUENCE OF ELEVATED INITIAL TEMPERATURE ON HYDROGEN COMBUSTION

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ABSTRACT

The effect of initial temperature on turbulent flame acceleration and deflagration - detonation transition (DDT) was investigated experimentally in an explosion tube (length 6 m, diameter 66 mm) with various obstacle courses in the flame path. The experiments were performed at initial temperatures between 80°C and 300°C with H₂/air/steam mixtures containing 8 to 20% H₂ and up to 55% steam by volume. Flame speed measurements indicate that the mixture ranges allowing a turbulent flame acceleration and a DDT become wider as the initial temperature is increased. Depending on the geometric boundary conditions, however, also damping effects can come into force leading to lower flame speeds in a given mixture.

INTRODUCTION

During postulated loss-of-coolant accidents in a light water reactor (LWR) large quantities of hydrogen can be generated by oxidation of the zirconium equipment and by core-concrete interaction following a failure of the pressure vessel. The hydrogen released may form a combustible mixture with the air and vapour in the containment. This mixture can be ignited by any ignition source present in the containment under accidental conditions such as hot surfaces, electric discharge etc.. Investigations as to whether combustion of the hydrogen possibly exerts pressure loads on the containment structure endangering its

integrity presently constitute an important sector in reactor safety research. Discussing the danger potential of pressure loads caused by hydrogen combustion in a reactor containment only premixed flames are of interest. The comparatively slow heat release of a more or less stationary diffusion flame imposes no particular danger on the integrity of the containment. Basically two kinds of combustion can be distinguished under premixed conditions: deflagration and detonation. In the containment only weak ignition sources initiating a deflagration are to be expected. Ignition sources of sufficient strength to start a detonation directly are not present. An initially slow deflagration, however, can be accelerated to very high flame speeds or even undergo a transition to a detonation (DDT) under the influence of turbulence inducing obstacles in the flame path, e.g. bundles of pipes, openings between two adjacent rooms.

Detonations as well as very fast deflagrations are characterized by a strong leading shock wave which can possibly endanger the integrity of the containment. So, detailed knowledge of the phenomena of turbulent flame acceleration and DDT is necessary for assessing the danger potential of the released hydrogen in the course of a core melt accident in a LWR.

Basically flame speed depends on the following parameters: mixture composition, geometry of confinement, initial thermodynamic conditions. Up to now research work was focussed on the influence of various geometries on the combustion process and its dependance on the mixture's composition. Many experi-

ments were conducted at initial temperatures between ambient temperature and about 100°C e.g. Peraldi et al. (1986)/1/, Sherman et al. (1989)/2/, Mayinger, Brehm (1988)/3/, Kumar, Bowles (1991)/4/, Chan (1991)/5/. In the course of a severe accident higher initial temperatures have to be expected. This paper presents investigations of the combustion behaviour of hydrogen - air - steam mixtures at elevated initial temperatures between 80 and 300°C .

THEORETICAL CONSIDERATIONS

Before presenting the experimental results, the effect of elevated initial temperature on a premixed combustion process occurring in a closed volume is briefly discussed theoretically. The aspects of adiabatic isochoric combustion, detonation, DDT, turbulent flame acceleration are controlling factors for the pressure loads imposed on the containment structures.

Pressure Ratio of Adiabatic Isochoric Combustion

In case of a comparatively slow deflagration of a given mass of a combustible mixture in a constant volume the pressure of adiabatic isochoric combustion forms an upper limit for the pressure built up. Treating the components involved as ideal gases the pressure p_2 after the combustion can be written as follows:

$$p_2 = p_1 \cdot \frac{R_2}{R_1} \cdot \left(1 + \frac{T_2 - T_1}{T_1}\right) \quad (1)$$

subscript 1: initial conditions, properties of unburnt gases

subscript 2: conditions, properties of burnt gases

R_1, R_2 : gas constants of unburnt and burnt mixture, respectively

For a rough approach dissociation and the temperature dependence of the specific heats can be neglected. With this assumption the temperature rise ($T_2 - T_1$) and the ratio R_2/R_1 are independent of the initial temperature T_1 . So it can be seen from equation (1) that the pressure ratio of an adiabatic isochoric combustion decreases as the initial temperature T_1 is increased. The increase of the heat capacities and the dissociation effects at higher temperatures enhance the described effect. Taking into account that the initial pressure p_1 of a given mass of gas in a given volume is proportional to the initial temperature T_1 , the pressure of an adiabatic isochoric combustion p_2 shows a comparatively weak increase

when raising the initial temperature.

Detonation

The pressure ratio at the leading shock wave of a detonation decreases with increasing initial temperature, thus showing a behaviour similar to the pressure build up of adiabatic isochoric combustion. This tendency can easily be noticed when considering a one dimensional detonation and treating the combustion process as an addition of heat to a flow of ideal gas with constant specific heat c_p . The Chapman Jouguet Mach number Ma_{CJ} and the pressure ratio of the leading shock wave can be calculated:

$$Ma_{CJ} = \sqrt{1 + \frac{\kappa + 1}{2} \frac{q}{c_p T_1}} + \sqrt{\frac{\kappa + 1}{2} \frac{q}{c_p T_1}} \quad (2)$$

$$\frac{\hat{p}_1}{p_1} = 1 + \frac{2\kappa}{\kappa + 1} (Ma_{CJ}^2 - 1) \quad (3)$$

κ : ratio of specific heats

Assuming the effective heat of reaction per kilogram q being constant for a given mixture it can be seen that the CJ-Mach number and thus the pressure ratio of the incident shock \hat{p}_1/p_1 decreases as the initial temperature T_1 is increased. Considering the rise of initial pressure p_1 , if the temperature T_1 is raised in a closed volume containing a given mass of gas, the superposition of both effects leads to an only weak increase of the detonation peak pressure for a given mixture.

The leading shock wave of a detonation, however, is not simply planar, but it shows a three dimensional structure due to smaller waves running across the shock front. The triple points of these secondary shocks can be visualized by means of a sooted surface arranged parallel to the direction of the detonation path. They draw a cellular pattern with a characteristic length scale onto the sooted surface. The length scale is denoted as detonation cell width λ , which forms an important parameter for estimating the detonation hazard of a given mixture: the smaller λ , the higher the probability of a detonation. In the temperature range mentioned here, calculations of Stamps et al. (1991) /6/ predict a decrease of λ with increasing initial temperature for fuel-lean hydrogen - air - steam mixtures. Mixtures near stoichiometry show a weak increase of λ due to dissociation effects at the very high temperatures reached in the reaction zone.

DDT

For the occurrence of a DDT a formation of a shock wave strong enough to cause self-ignition of the given mixture is necessary. Such a shock wave may be generated, for instance, by a local explosion in the vicinity of the reaction zone of an already highly accelerated deflagration or by shock focussing depending on the boundary conditions. With an increase of the initial temperature of a given mixture the temperature rise of a shock wave necessary for self-ignition becomes smaller and thus the minimum pressure ratio of a shock wave necessary to initiate a DDT decreases. So an increase of the initial temperature facilitates the occurrence of a DDT.

Turbulent Flame Acceleration

The mixture range allowing a strong flame acceleration is expected to become wider with increasing initial temperature for two reasons: For one, the laminar flame velocity for a given mixture increases with a raise of the initial temperature. Therefore, a high flame speed can be reached with a smaller extent of turbulence induced folding of the flame front. Second, the quenching effects due to turbulent mixing of cold unburnt and burnt gas in the flame front have to be considered. These quenching effects are the reason for flames in fuel lean mixtures not being able to reach high flame speeds. With increasing initial temperature, however, the quenching effect of the unburnt gas becomes smaller.

The possibility of a flame actually reaching a maximum flame speed specific for a given mixture highly depends on the turbulence induced to the flow. An increase of the initial temperature leads to a decrease of the expansion ratio between burnt and unburnt mixture, as can be seen from the temperature behaviour of the adiabatic isochoric pressure ratio discussed above. So the gas motion of the unburnt gas ahead of the flame front caused by the expansion of the hot gas behind the flame front becomes slower at the same time. Therefore, the obstacles in the flow field induce less turbulence in the unburnt mixture ahead of the flame front. Thus, an inhibiting effect of elevated initial temperature on turbulent flame acceleration due to the effect on the flow field has to be expected. With the current understanding of turbulent flame acceleration it is not possible to decide which of the two effects – the enhanced reactivity or the damping effect of the flow behaviour – dominates under any given conditions (mixture, geometry,

initial thermodynamic conditions).

EXPERIMENTS

The main objective of the experimental investigations was to determine the mixture limits for a strong turbulent flame acceleration in H₂-air-steam mixtures with the initial temperature as parameter.

Flame Speed Measurements in an Explosion Tube

The experiments were conducted in an explosion tube, closed at both ends, with a length of 6 m and a diameter of 66 mm. To achieve a uniform temperature distribution the tube was encompassed with an annulus filled with heat carrier oil that is heated up to the desired initial temperature by means of an electric heating wire. The maximum temperature was limited to 300°C due to the temperature range of the heat carrier oil. A defined hydrogen-air-steam mixture in the tube is ignited by means of a spark plug fixed on one of the end flanges of the tube. After ignition the flame propagation is recorded by a system of photodiodes installed in equidistance of 40 cm over the entire length of the tube (fig. 1). Various obstacle courses were employed as turbulence generators. The obstacle courses were made of orifice rings.

For the determination of the mixture limits for turbulent flame acceleration a geometric configuration of the obstacle course is required that causes a maximum turbulent flame speed for any mixture and initial condition. In former experiments conducted at the same facility the combustion behaviour of H₂-air mixtures under initial conditions of 80°C temperature and 1 bar pressure was investigated employing various obstacle courses as turbulence generators. Orifices of two different blockage ratios (free cross section of the orifice related to tube cross section) BR=0.7 and BR=0.32 (fig. 2) were used. The spacing between obstacles was varied between 50 and 500 mm. The obstacle courses had a length of 3 m beginning at the ignition end of the tube. The remaining 3 m were free of obstacles.

In obstacle courses comprising orifices with the high blockage ratio the highest flame speeds were observed for all mixture compositions, if the orifices were arranged at a regular spacing of 500mm. A smaller spacing caused a higher initial acceleration after ignition. The maximum flame speeds, however, remained lower due to pressure losses at the obstacles. In contrast to that, the maximum effect on flame acceler-

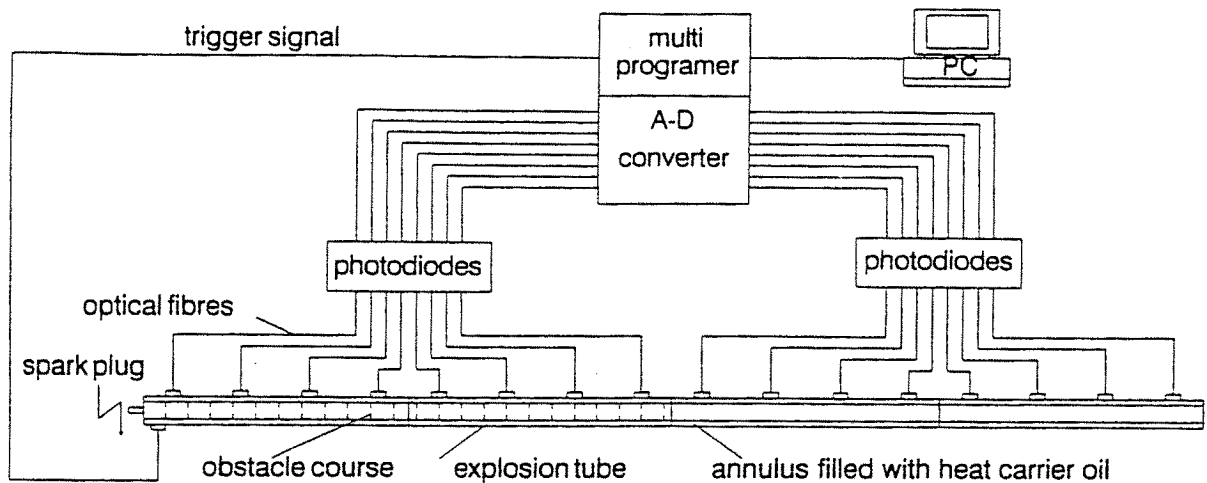


Fig. 1: Explosion tube with photodiode system for flame speed measurements

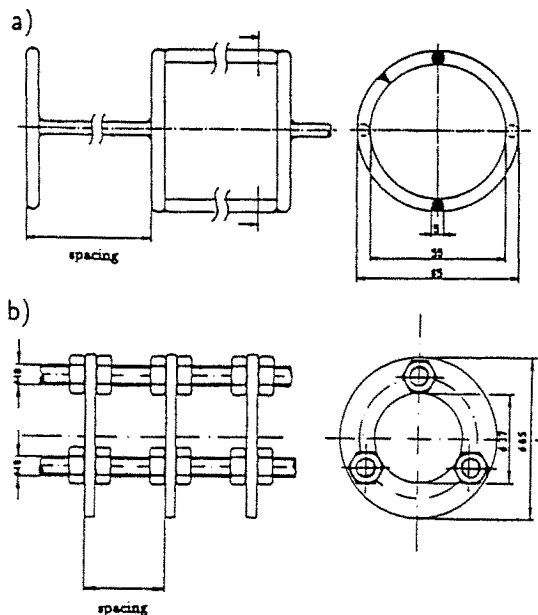


Fig. 2: Obstacle courses used for combustion experiments in the explosion tube: a) turbulence inducing obstacles, blockage ratio 0.3, b) jet stream promoting obstacles, blockage ratio 0.7

ation was achieved with the obstacles of the smaller blockage ratio when being mounted at a very small spacing of 50 mm. With this type of obstacles the same maximum flame speed for each mixture was observed as in the most critical obstacle course with the high blockage ratio. The acceleration process, however, was slower, in other words, the distance from the ignition source where a maximum flame speed

was reached was longer. Particularly fuel lean mixtures reached a maximum flame speed very close to the end of the obstacle section. The influence of the spacing of the obstacles being different for the two types of orifices with different blockage ratios, can be explained by the way turbulence is induced in each case: the orifices with the small blockage ratio have an effect on the flow similar to a rough wall, whereas an orifice with a high blockage ratio causes the formation of a highly turbulent jet through the opening with the effect of a very fierce acceleration of the flame. Thus only a few orifices suffice to reach very high flame speeds.

In order to make sure that a mixture specific maximum flame speed is reached in the tube, additional tests were conducted with a third obstacle arrangement that causes a stronger turbulent flame acceleration than the two courses described above. Keeping in mind the described effects of the blockage ratio of the orifices on the combustion process, this third arrangement was designed in the following way: the first meter of the tube, beginning at the ignition end, was supplied with 7 orifice rings with a blockage ratio of $BR = 0.7$ mounted in equidistance of 140 mm, followed by orifices of the small blockage ratio $BR = 0.3$ with a constant spacing of 50 mm over a length of 3 m. So a very strong acceleration is achieved in the beginning of the flame path by the orifices with the high blockage ratio. In the second section of the obstacle course a further acceleration of the flame front is possible, since the losses of a pressure wave travelling ahead a very fast deflagration front are kept small and a high degree of turbulence is induced to the

flow at the same time. The maximum flame speeds reached in the third obstacle course were the same as observed in the two most effective courses tested before. For these three most critical obstacle courses described the lean limit for a strong flame acceleration was 10% H₂ in a H₂/air mixture. This value is in good accordance with results obtained in other small and large scale facilities at initial temperatures between ambient temperature and about 100°C by other researchers. So 10% H₂ in H₂-air obviously forms the lower limit for a strong flame acceleration being possible at the initial conditions considered here. On the other hand, it can be concluded that the three obstacle configurations mentioned here cause a mixture specific maximum flame speed.

So these three obstacle arrangements were employed for the combustion tests at elevated initial temperature with the aim of determining mixture limits for a strong turbulent flame acceleration in H₂-air-steam mixtures. The initial partial density of the air in the mixture was kept at a constant value equal to 1 bar and 40°C corresponding to thermodynamic states in a real containment. The total initial pressure, however, had to be limited to 2 bars, to avoid pressure loads on the facility beyond the design pressure in case of detonations occurring. The first two test series were conducted with the following obstacle courses already described above: series 1: orifices with a blockage ratio of 0.3 mounted in equidistance of 50 mm, series 2: orifices with a blockage ratio of 0.7 mounted in equidistance of 500 mm.

Results are shown in Fig. 3. For both obstacle courses the lean limit for a strong turbulent flame acceleration is shifted towards higher values of H₂ content as the initial temperature is increased. The mixture range for a DDT occurring in the tube, however, widens at the same time. The lean limit for a DDT was observed at 12.5% H₂ in series 1 and 17% H₂ in series 2 in dry H₂-air mixtures at 200°C in contrast to 20% H₂ for both series at 90°C. The very high flame speeds near CJ-velocity at 90°C initial temperature in mixtures with less than 20% H₂ are caused by very fast deflagrations, not by a detonation. They can be distinguished by the fact, that the speed of a deflagration drops after the flame has left the obstacle course due to the lack of turbulence inducing obstacles, whereas a detonation shows an increase of the speed of the reaction front because the pressure losses in the obstruction free tube section are smaller. Only small quantities of steam in the mixture were sufficient to make a DDT impossible at any initial

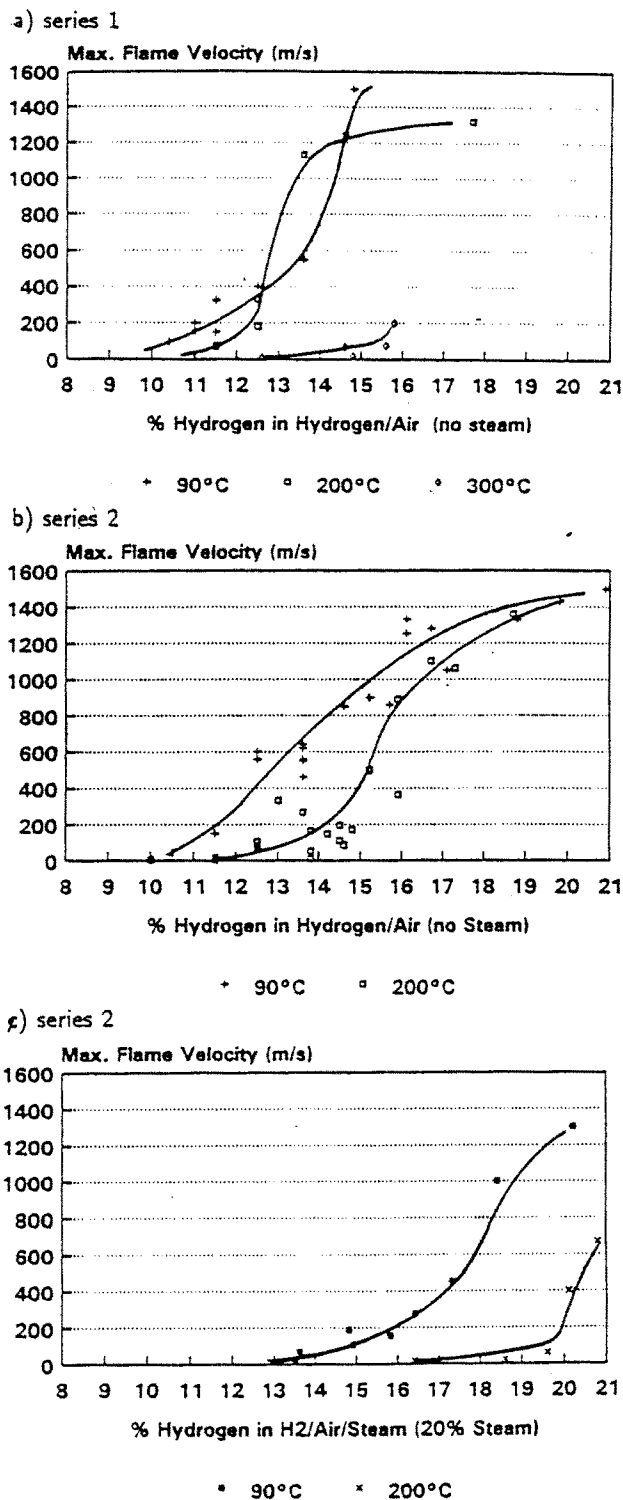


Fig. 3 Maximum flame speeds recorded in the explosion tube, a) test series 1: obstacle course: length 3m beginning at ignitor, blockage ratio 0.3, spacing 50 mm, b),c): test series 2: obstacle course: length 3m beginning at ignitor, blockage ratio 0.7, spacing 500 mm

temperature and H₂-concentration. Additional tests at 300°C in series 1 only yielded a very weak flame acceleration.

In the third test series in the explosion tube the third obstacle course described above comprising two different types of orifice rings arranged in a row of 4 m in length served as turbulence generators. The combustion behaviour of mixtures containing between 8 and 21% H₂ and between 0 and 55% steam were investigated at initial temperatures of 100, 200, 280°C. The maximum flame speeds measured are summarized in Fig.4. The initial temperature has the following effects on the combustion process: For dry mixtures a rise of the initial temperature results in a decrease of the lean limit for a strong turbulent flame acceleration: 10.5% H₂ at 100°C, 9% at 280°C were observed in the tests. The lean limit for a DDT also drops: 20% H₂ at 100°C, 12.5% at 200°C, 12% at 280°C. In mixtures containing steam no DDT was recorded at 100°C initial temperature. In mixtures with a small steam content of 7% or less and a H₂-content of more than 14.5% a DDT occurred at 200°C. At 280°C in a mixture with a steam concentration of 15% and a H₂-content of 16.5% a DDT was observed. For steam concentrations higher than 15% no DDT occurred in the tests.

In mixtures with a steam content up to 30% a rise of the initial temperature leads to an increase of the maximum flame speed recorded in the tube for a particular mixture. At higher steam concentrations the enhancing effect of elevated initial temperature could not be detected. The upper limit of the steam content for a strong turbulent flame acceleration was 50% at all initial temperatures tested. Mixtures with more than 53% steam could not be ignited at all initial temperatures tested.

Flow Measurements in Explosion Tube

In the first two test series of the flame speed measurements conducted in the explosion tube a weaker turbulent flame acceleration was observed when raising initial temperature. As already mentioned in section (2), this effect has to be explained by the decrease of the expansion ratio between burnt and unburnt mixture having a damping influence on gas motion and turbulence generation. This effect was proved experimentally by flow measurements in the combustion induced gas motion employing a Laser - Doppler - Velocimetry (LDV) system. A scheme of the measuring arrangement is given in Fig.5. Time based pro-

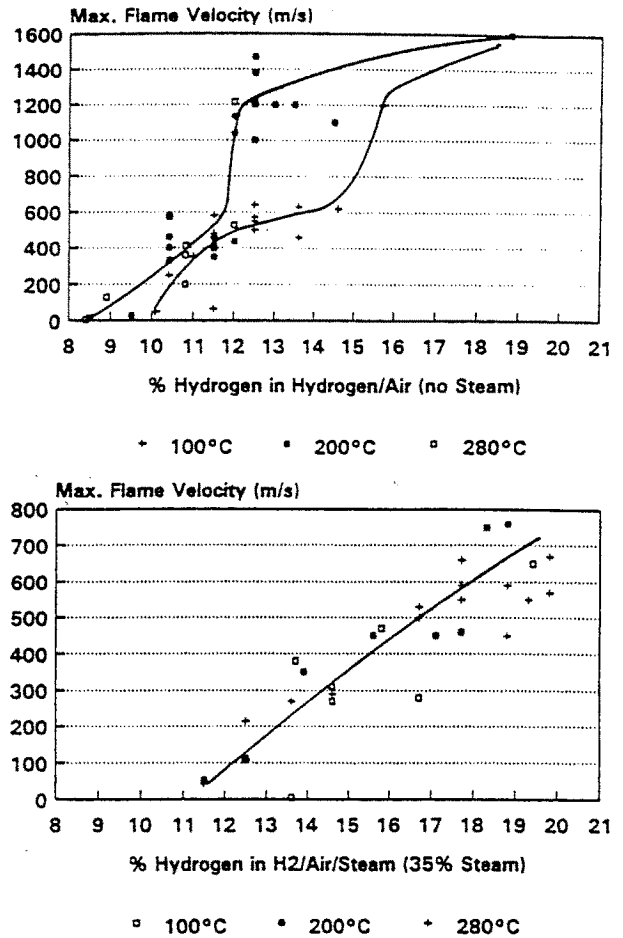


Fig. 4: Maximum flame speeds recorded in the explosion tube, test series 3: obstacle course: 4m beginning at ignitor: 1m with blockage ratio 0.7, spacing 140 mm followed by 3m with blockage ratio 0.3, spacing 50 mm

files of the main flow velocity were recorded. The measuring volume was adjusted to the center of the tube cross section, 1.6m from the ignitor. The LDV-system comprised a forward scatter setup with the following features: A 3 W argon-ion laser served as light source. The LDV optics was equipped with a Bragg-cell for frequency shift facilitating a measuring range between 250 m/s in direction of flame propagation and 400 m/s in the opposite direction. The Doppler frequencies were detected by a counter. TiO₂ powder was used as scattering particles.

For the flow measurements the obstacle course used in the first test series of the flame speed measurements was employed (orifice rings with BR=0.3 mounted at equidistance of 50 mm). H₂-air mixtures with a H₂-content of 12 and 14% were used for the

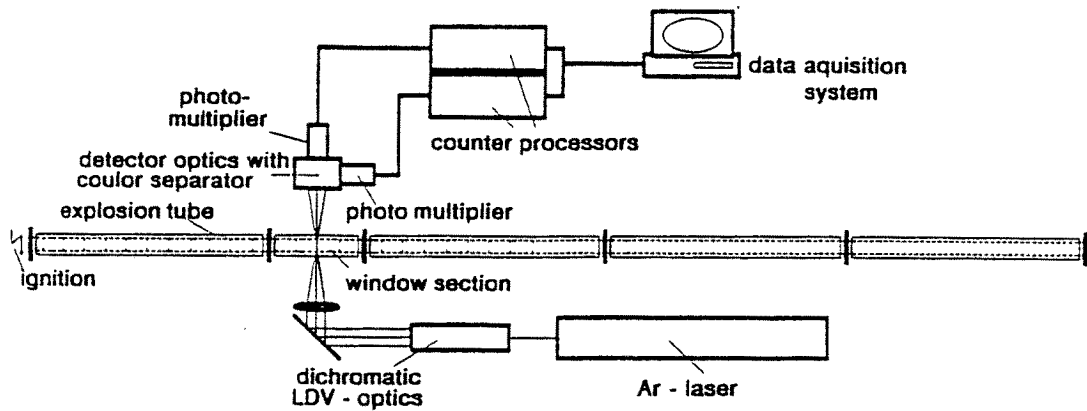


Fig. 5: Explosion tube: Setup of Laser - Doppler - Velocimeter

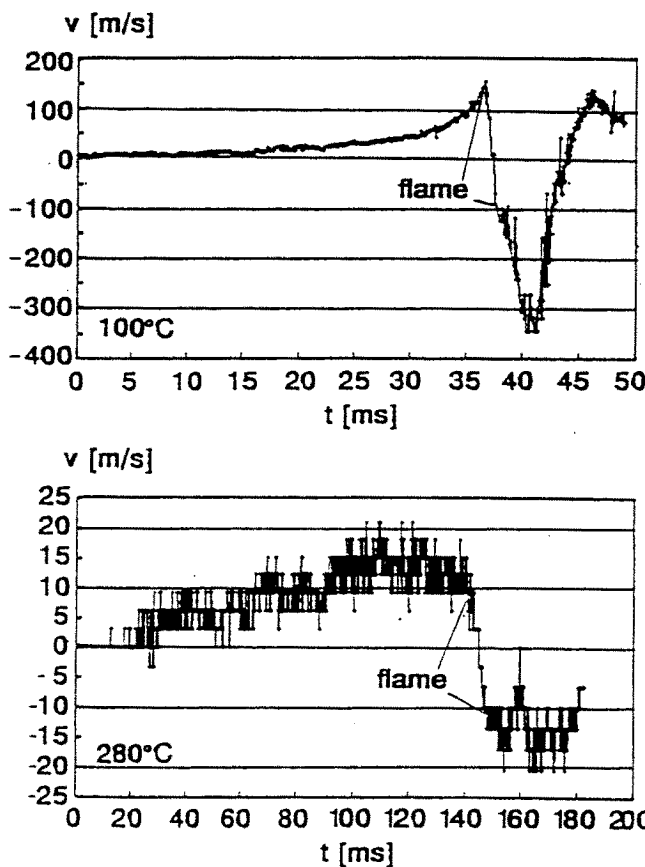


Fig. 6: Explosion tube: time based velocity profiles of combustion induced gas movement, H_2 -concentration 12% in H_2 -air, initial temperatures $100^\circ C$ and $280^\circ C$, obstacle course: length 3m, blockage ratio 30%, spacing 50mm

test runs. The influence of the initial temperature on the combustion induced gas motion was tested at initial temperatures of $100^\circ C$ and $280^\circ C$. The load

of combustible mixture was kept constant for both temperatures by setting the initial pressure to 1 bar and 1.4 bars, respectively. Basically the time based profiles of the gas velocity (see fig. 6) show the following behaviour: After ignition the unburnt mixture ahead of the flame front moves at an increasing velocity in direction of the flame propagation. When the flame passes the measuring volume the flow direction is reversed within a few milliseconds due to the heat addition in the reaction zone.

Comparing the velocity profiles obtained at $100^\circ C$ and $280^\circ C$ initial temperature it can be seen that the velocities of the gas motion at the higher temperatures are much lower, thus showing the described effect.

Tests in a stationary tube type burner

Within the temperature range mentioned in this paper the reaction rate is increased as the temperature is increased. This effect was investigated in a stationary tube type burner in which stationary flames have been stabilized behind metal grid obstacles. Fig.7 shows the minimum H_2 -concentration required to achieve a stable flame behind a grid with a blockage ratio of 55 % above the approach velocity of the unburnt gas for different temperatures. For stationary flames the approach velocity of the unburnt gas is equal to the overall burning velocity. It can easily be seen that the required H_2 -concentration decreases with the increasing initial temperature. Assuming constant flow conditions at constant approach velocity it can be deduced, that quenching effects decrease with increasing initial temperature. Measurements of the length of the reaction zone, conducted in

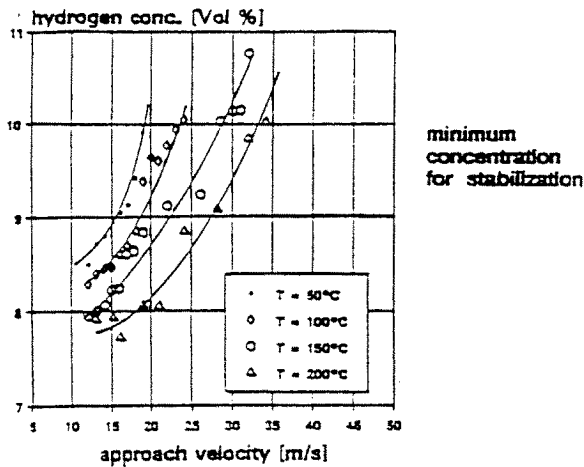


Fig. 7: Minimum H₂-concentration required for a stable, stationary highly turbulent flame behind a metal grid (blockage ratio 55%) as a function of approach velocity of the unburnt gas and initial temperature.

the same burner, exhibited a decrease in the length of the reaction zone with increasing initial temperature also indicating a higher reactivity in the gas.

DISCUSSION OF RESULTS

The three obstacle configurations investigated in the explosion tube lead to a mixture specific maximum flame speed at initial temperatures up to 100°C. At higher temperatures only the third obstacle course comprising two different types of orifices lead to the widening of the mixture range for a strong turbulent flame acceleration that has to be expected due to enhanced reactivity and diminishing quenching effects. For mixtures containing no steam a decrease of the lean limit of the H₂-content for a strong flame acceleration from 10.5 to 9.5% was observed between 100 and 200°C and down to 9% at 280°C. There are no experimental data available from other researchers up to now to compare these results. So it is difficult to judge for initial temperatures of more than 100°C why no flame acceleration is observed in mixtures below the lean limits mentioned above from the tests in the explosion tube. There are two reasons possible for a turbulent flame acceleration not to occur in a given mixture. Either quenching effects inhibit the acceleration process. In this case the mixture composition lies outside the range for a turbulent flame acceleration being possible. Or not enough turbulence is induced to the flow, due to the turbu-

lence generators not being effective enough to cause a considerable flame acceleration. So the experiments showed that an increase of the initial temperature of a given mixture can have an enhancing or damping effect on turbulent flame acceleration depending on the geometric boundary conditions and as a result of this that the influence of the geometry on turbulent flame acceleration gains importance with increasing initial temperature.

At the same time a DDT can occur with a smaller extent of turbulent flame acceleration preceding the transition and the mixture range for the possibility of a DDT becomes wider. At 280°C a fully developed detonation was observed in a H₂-air mixture with only 12% H₂.

When applying the results obtained in the small scale facility to a real containment scaling effects play an important role. Due to the lack of knowledge on the field of turbulent combustion only making possible qualitative predictions of flame acceleration processes it cannot be judged by what extent the inhibiting effects of elevated initial temperature on turbulent flame acceleration come into force in large scale geometries. Considering the phenomena of DDT and detonation it has to be taken into account that a detonation requires a minimum channel width of the order of the detonation cell-width λ to be sustained. Consequently less reactive mixtures, i.e. mixtures with a higher steam content, or fuel leaner mixtures than in the explosion tube suffice to make possible a DDT in a larger geometry. Thus a conservative assessment of the influence of elevated initial temperature on a hydrogen combustion process in a LWR containment leads to the conclusion that the danger potential of a given mixture under given geometric boundary conditions increases as the initial temperature increases.

CONCLUSIONS

The experiments show that on the one hand the mixture potential for a turbulent flame acceleration increases when the initial temperature is raised, but on the other hand more effective turbulence inducing obstacles in the flame path are necessary to activate this potential. The mixture range for a DDT widens at the same time.

To reduce conservatism of the assessment of the danger potential the knowledge about the interactions between flowfield and reaction zone being the key for a quantitative prediction of a combustion process, has to be improved.

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