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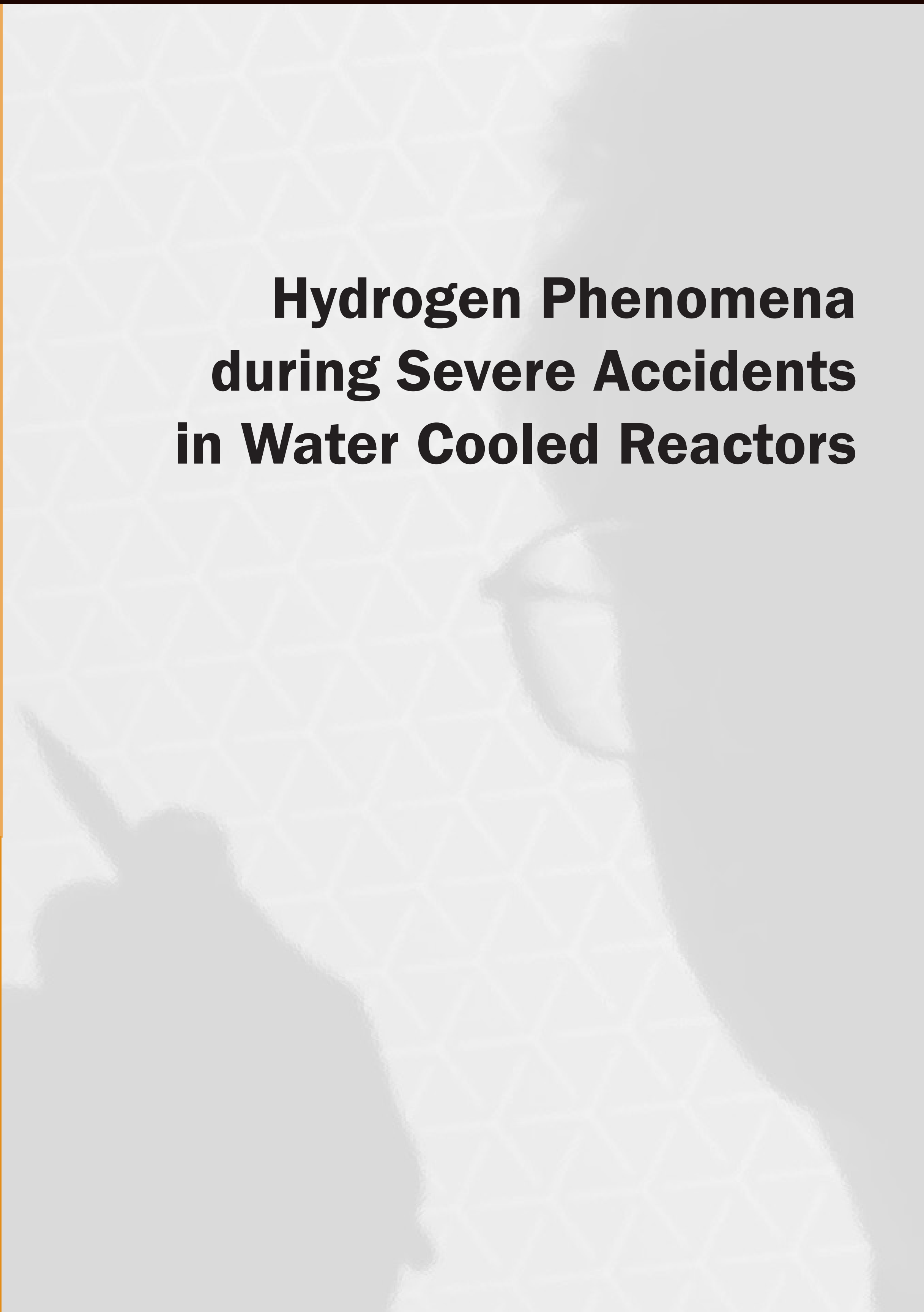

Hydrogen Phenomena during Severe Accidents in Water Cooled Reactors

VIENNA, 2021

TRAINING COURSE SERIES 72

HYDROGEN PHENOMENA DURING SEVERE ACCIDENTS IN WATER COOLED REACTORS

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INTERNATIONAL ATOMIC ENERGY AGENCY
VIENNA, 2021

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HYDROGEN PHENOMENA DURING SEVERE ACCIDENTS IN WATER COOLED REACTORS
IAEA, VIENNA, 2021
IAEA-TCS-72
ISSN 1018–5518

Printed by the IAEA in Austria
February 2021

FOREWORD

The IAEA offers its Member States a wide spectrum of education and training activities. These include face-to-face training courses and workshops, on-line learning, fellowship programmes and schools, as well as publications in the IAEA's Training Course Series, including handbooks, textbooks and manuals on various nuclear related topics.

The IAEA shares the latest knowledge and findings with its Member States on the topics of most interest to support further development and sustainability of national nuclear power programmes. This Training Course Series publication is aimed at providing substantive details of hydrogen behaviour fundamentals with sufficient background information to enable readers to understand generation, propagation, combustion and mitigation of hydrogen during severe accidents in water cooled power reactors. Examples demonstrating the theory fundamentals are included. This introductory text can be used to gain a conceptual understanding of the relevant phenomena to build foundational knowledge for further education on hydrogen behaviour during severe accidents. The content is closely linked with IAEA-TECDOC-1939, Developments in the Analysis and Management of Combustible Gases in Severe Accidents in Water Cooled Reactors following the Fukushima Daiichi Accident.

This information included here can be applied directly in training courses, as lecture material or as student or trainee reading/reference material, and incorporated into the curricula on related nuclear topics. The publication is expected to be of benefit to students or trainees as a basic reference text, where the basic concepts covered are intended to directly support the analyses of hydrogen behaviour during severe accidents in water cooled power reactors.

The IAEA acknowledges the contributions of S. Gupta (Germany) and A. Bentaib (France) in the creation of this publication. The IAEA officer responsible for this publication was T. Jevremovic of the Division of Nuclear Power.

EDITORIAL NOTE

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CONTENTS

1. INTRODUCTION

1.1. BACKGROUND

There has been an increase in interest among Member States for education and training on severe accident phenomena, with hydrogen-related phenomena being of particular interest following the Fukushima Daiichi nuclear power plant (NPP) accident in 2011. Hydrogen behaviour during a severe accident in water cooled power reactors is complex process, and the related analysis, management and mitigatory measures rely on a thorough and informed understanding of related hydrogen phenomena.

Education and training on nuclear energy, nuclear reactor technologies, nuclear systems, reactor components and nuclear safety features are of great importance. In order to support the development of education and training courses, this publication serves as a general resource for information on hydrogen as it relates to water cooled reactor operation and severe accidents conditions.

This publication is intended to provide users with a ‘big picture’ and basic principle understanding of hydrogen generation, propagation, combustion and mitigation techniques of relevance to water cooled power reactors. It is intended to serve as a basis of knowledge that can be applied to general nuclear engineering education. The publication is therefore valuable to students or trainees as a basic reference text where the basic concepts covered are intended to directly support the analyses of hydrogen behaviour during severe accidents in water cooled power reactors.

This publication can be used for education and training programmes including universities and technical training institutes among others. The contents may be integrated into educational and training courses focused on hydrogen in NPP containment or as a supplement to courses with a broader, related curriculum.

1.2. OBJECTIVE

The objectives of this publication are to:

- Provide comprehensive information on hydrogen properties, its generation, distribution, combustion and mitigation in water cooled power reactors during severe accidents;
- Provide a resource for development of training workshops and training courses on hydrogen behaviour during severe accidents;
- Support the development of educational and training curriculum in universities and technical training institutes by providing an overview of hydrogen behaviour as relevant to NPPs;
- Serve as a reference for lectures on basic theory and fundamentals on hydrogen behaviour in water cooled reactors;
- Serve as a reference for students and trainees, who may be unfamiliar with hydrogen behaviour during severe accidents in water cooled power reactors (e.g. engineering

safety, physical sciences, non-nuclear engineering fields, and including non-technical fields) to understand the theory at a basic principle level.

1.3. SCOPE

The scope of this publication is to present information on hydrogen behaviour and management in water cooled reactors during severe accidents focusing at descriptions on hydrogen properties, its generation, distribution and combustion. The publication includes an overview of hydrogen risks, analysis, measurement, and mitigation strategies. The level of explanation is restricted to conceptual knowledge and basic principles of hydrogen behaviour and includes relevant examples and quiz questions.

The overall scope of the publication is developed to support training workshops and education programmes in Member States.

1.4. STRUCTURE

This publication contains the following sections:

- **Physics and Chemistry of Hydrogen** provides an overview of hydrogen properties relevant to later explanations on its behaviour in water cooled reactors during severe accidents.
- **Hydrogen Behaviour in Water Cooled Reactors** provides background information on relevant nuclear principles on hydrogen generation and propagation in water cooled reactors during severe accidents and discusses the risks of hydrogen combustion.
- **Mitigation Measures** provides an overview of the systems and mitigation techniques to remove and monitor hydrogen propagation within and outside of the containment.
- **Review Questions and Exercises** contains quiz-type questions and examples with explanations and detailed discussions.

2. PHYSICS AND CHEMISTRY OF HYDROGEN

2.1. HYDROGEN PROPERTIES

Hydrogen gas, with molecular weight of 2.016 g/mol, is lighter than other gases such as steam, carbon monoxide, carbon dioxide and air (mostly consisting of oxygen and nitrogen) which are all present inside the NPP containment during a severe accident. Thus, hydrogen gas is likely to be found in the upper part of the containment due to the buoyancy force.

The dynamic viscosity of hydrogen at 0°C is 8.4×10^{-9} Pa·s. It is lower than the dynamic viscosity of saturated steam at 1 bar which is 12×10^{-9} Pa·s and lower than half of the dynamic viscosity of air at 0°C which is 17.4×10^{-9} Pa·s. This means that when hydrogen gas moves upward, the air entrainment would be more intense than the steam entrainment[1].

The diffusivity of hydrogen and steam in the air at temperature of 25°C and pressure of 1 atm (101.33 kPa) is 0.41×10^{-4} m^2/s and 0.26×10^{-4} m^2/s respectively. Thus, the diffusion of hydrogen is expected to be greater than that of steam, causing formation of the regions with high hydrogen concentrations [1].

2.2. HYDROGEN MIXING

The mixing process is the phenomenon when multiple fluids with different characteristics come together and become a single fluid with one characteristic. It is an intra-compartmental process which includes all phenomena in a single compartment whereas the transport process is an inter-compartmental process driven by pressure difference or convection. In case of large pressure difference between upstream and downstream regions, hydrogen transport by pressure difference will dominate all other mechanisms. The mixing process can be driven by forced or natural convection or at much slower rate by diffusion. Stratification may occur when the mixing process is incomplete.

When hydrogen flows and diffuses in the air–steam atmosphere of the containment, it is mixed with the air–steam mixture and the air–steam–hydrogen (or other combustible gases e.g. carbon monoxide) mixture cloud, which may form in some regions of the containment for some time. If hydrogen containing gas-mixture release has initial momentum and higher temperature than surroundings, some of its energy is transferred to surrounding gases but eventually accelerates the separation of hydrogen from other gases. In the extended absence of additional driving forces hydrogen gradually separates, moves upward and accumulates in the upper regions of the containment, a phenomenon known to be stratification. Therefore, the mixing process is an important phenomenon because it decides how gases such as hydrogen can distribute and accumulate inside the containment.

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[1] Entrainment is the transport of fluid across an interface between two bodies of fluid by a shear induced turbulent flux.

The transport and mixing of hydrogen inside the containment of light water reactors (LWRs) are critical in determining the time and mode of hydrogen combustion. Rapid mixing could result in uniform distribution of hydrogen. Slow mixing may lead to localized burning and locally detonable mixtures. Gas atmosphere mixing is a very important phenomenon in nuclear reactor containments during the severe accidents. The evaluation of structural integrity includes the assessment of pressure loads due to release of steam and gases and the combustion of hydrogen. Since hydrogen combustion is highly dependent on the local concentration ratios, the propagation behaviour of hydrogen in the containment atmosphere becomes important.

For the control of hydrogen propagation there are a number of devices and measures in the NPPs. In order to assess the risk of energetic hydrogen combustion, accurate prediction of atmospheric distributions of gas components, steam, temperature, as well as flow and mixing mechanisms are required. The gas concentration is lowered by natural convection and turbulent diffusion. These processes tend to reduce a potential for energetic combustion within the containment. The passive and active hydrogen recombination presenting mitigation measures are affected as well by the hydrogen concentration and its distribution within the containment volume.

What is the role of forced convection, natural convection and diffusion: The mixing processes are affected by the rate of hydrogen released and its distribution in the presence of steam (two phase effect) into the containment. The physical processes which govern the mixing in gaseous mixtures are forced convection, natural convection and diffusion. Natural convection occurs due to density differences of gases in the containment. Convection motions occur as lighter gases rises on one side and heavier gases falls down on the other side. The temperature difference between containment walls and gases atmosphere also plays a significant role in driving the convection. Convection can also occur due to the presence of active systems, e.g. mixing fans, sprays, or passive mechanisms such as momentum dominated jets from pipe break or valves. Diffusion occurs due to concentration gradient but may be of less importance in case of an accident when either natural or forced convections are expected to prevail over diffusion, gases will tend to diffuse from regions of higher concentration to regions of lower concentration following the Fick’s law.

Why stratified atmosphere is unfavourable: If mixing does not proceed to completion, but flow stagnates, then a stratified condition will be created potentially contributing to the risk of early containment failure. For example, during the severe accident with core meltdown in the pressurized water cooled reactors (PWRs) that have containments with large volumes and the atmosphere that is not inert during normal operation, a release of hydrogen gas from the primary system may lead to a formation of high detonable gas concentrations in the upper dome of the containment. In the case of long lasting accidents, hydrogen enrichment may also occur in the lower compartments of containment (inverse stratification). A further analysis about modelling of iodine behaviour and venting lines with Venturi scrubbers for filtered containment venting of a KONVOI type also showed an enrichment of hydrogen in the compartments which connect to the venting line [3]. Although both analyses indicated no increased risk of hydrogen deflagration inside the containment due to an almost complete oxygen consumption by the passive autocatalytic recombiners (PARs) in earlier accident phases, the latter emphasized a risk of deflagration or even detonation within the venting system and vent stack during the course of filtered containment venting under situations when hydrogen rich oxygen lean containment atmosphere is mixed with air.

The energetic combustion of hydrogen is expected at concentrations above 15% if the atmosphere is not inert by steam. The atmosphere becomes inert at volumetric steam concentrations above 55%, while for example in the International Standard Problem ISP-47 on Containment Thermal Hydraulics (ISP-47 THAI test) the steam concentration was below 20% [4]. From the measured and simulated gas concentration distributions, the detonable gas fraction can be determined by taking the detonation limit as hydrogen concentration of 16%. The results are shown in FIG. 1.

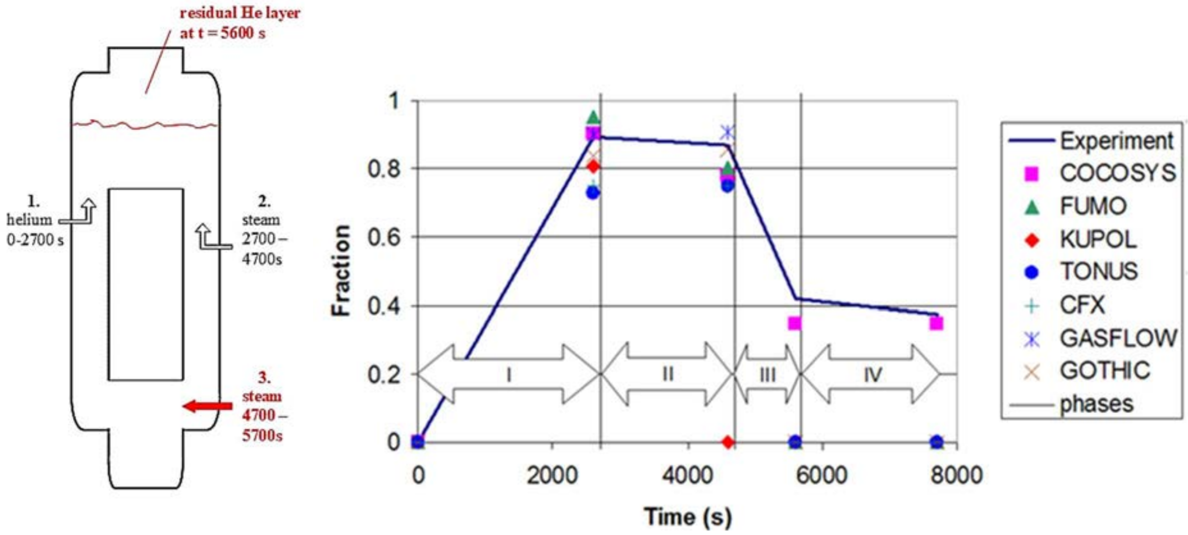

FIG. 1. *Experimental and simulated fractions of detonable gas mass [4].*

The experimental data as shown in FIG. 1 with initially stratified gas atmosphere and dissolution by gases released in lower part of containment allow assessing prediction capability of safety analysis tools to analyse locally stratified atmospheres in containments of NPPs.

What is affecting the mixing in containment atmosphere: Several parameters can influence the mixing behaviour of a containment atmosphere. Usually, forced convection involves higher gas flow velocities than natural convection. These higher velocities may induce enhanced mixing due to turbulence. The erosion rate is also affected by the jet diameter and velocity at the contact with the light-gas cloud edge, as well as by the density difference between the light-gas cloud and the lower layer. Turbulent waves are generated on the tip of the jet; the entrainment of hydrogen (being light gas) is determined by the breaking and overtopping of these waves. Therefore, the backflow of the hydrogen–air mixture occurs in an annular region around the rising jet; for example, FIG. 2 shows a vortex formation at the interface, similar to the flow of a vertical water fountain in the air.

The rising jet is subject to turbulent momentum and mass exchange with the annular downward flow. The mixing behaviour is significantly affected by density difference of stratified light gas layer and the penetrating fluid. The injected fluid travels through containment atmosphere before reaching the sharp density interface of the light gas. The flow is positively buoyant due to the injection of lower density fluid into higher density fluid. Jet flow may become negatively buoyant once it reaches the fluid layer containing light gas from upper layer and therefore, buoyancy forces and jet momentum starts to oppose one another. Due to the negative buoyancy, upwards flowing fluid will slow down until the buoyant jet reaches a maximum penetration height.

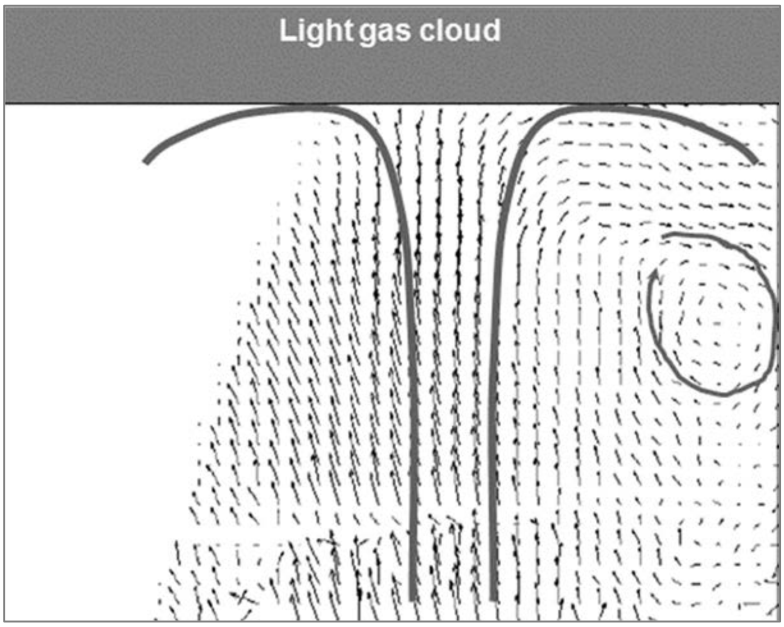

FIG. 2. Gas entrainment and vortex formation at the light gas layer interface [courtesy of Becker Technologies, Germany].

In studies of stratified layer dissolution and to facilitate analyses of different scenarios, non-dimensional numbers are generally used. The entrained hydrogen flow Q_{H2} can be expressed as a function of the impinging volumetric jet flow by means of the non-dimensional entrainment coefficient E:

$$Q_{\mathrm{H2}} = E \cdot Q_{\mathrm{jet}} \tag{1}$$

Baines [3] postulated that the entrainment can be expressed as a function of the Froude number or the Richardson number $Ri = Fr^{-2}$ as follows:

$$E = f(Fr) \tag{2}$$

It is important to mention that the Froude number needs to be calculated by physical quantities defined at the point of jet impingement:

$$Fr = \frac{u_{\mathrm{impg.}}}{\sqrt{r_{\mathrm{impg.}} \Delta_{\mathrm{impg.}}}} \tag{3}$$

where $u_{\text{impg.}}$ represents the jet velocity before the point of impingement and $r_{\text{impg.}}$ represents the half of the jet diameter at this location. The reduced gravity at the point of impingement $\Delta_{\text{impg.}}$ is defined as a function of the density of the jet ($\rho_{\text{jet @impg.}}$) and the density in the light gas stratification (ρ_{layer}) which creates the buoyancy force:

$$\Delta_{\text{impg.}} = g\frac{\rho_{\text{jet @impg.}} - \rho_{\text{layer}}}{\rho_{\text{layer}}} \tag{4}$$

Froude number can be calculated based on the spreading of a free jet scaling. For a free jet, self-preserving reason is reached at a certain distance away from the source. In the self-preserving region, jet spreading becomes independent from source conditions and is defined mainly by entrainment of a surrounding gas [5]. The correlation between entrainment coefficient and Froude number is given by correlation from Kumagai [6]:

$$E = \frac{Fr^3}{1+3.1\,Fr^2+1.8\,Fr^3} \tag{5}$$

The Kumagai correlation using free jet approach can be used for calculating the Froude number and comparison with experimental results, as shown in FIG. 3 [7]. This finding indicates that the quality of how exact the erosion of the light gas stratification can be estimated with the Kumagai correlation requires a precise knowledge of the jet or plume which is penetrating into the stratification layer.

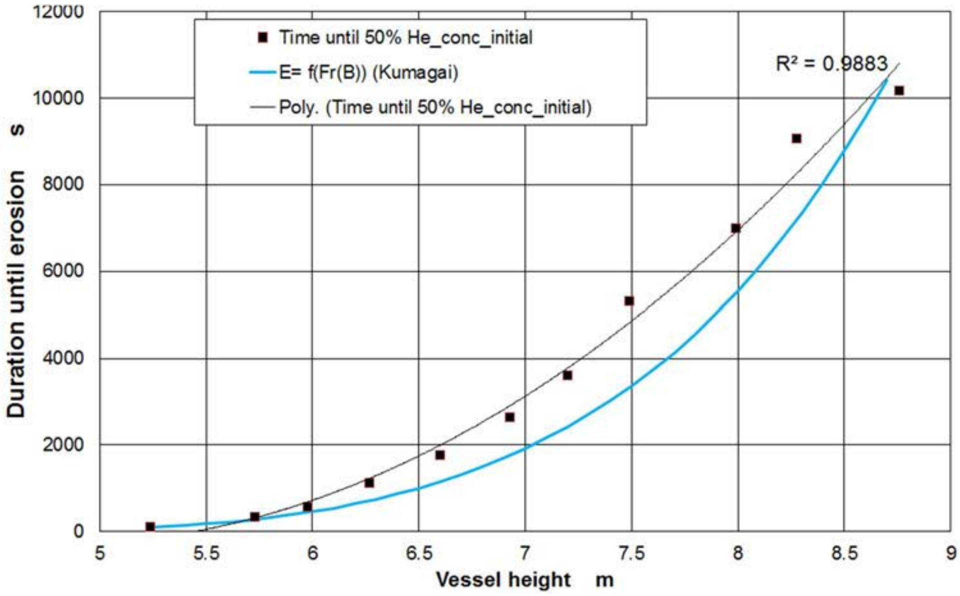

FIG. 3. Experimental and literature data on duration until erosion of the helium stratification as a function of containment vessel height [7].

Example of passive/active mixing: Outer surface cooling of containment vessel is presently considered in new rector design concepts, such as AP1000, as part of passive containment cooling system design. The concept is also proposed for the Japanese BWR designs as a voluntary accident management measure to prevent an over temperature damage of flange seal material [8]. The impact of steam condensation resulting from the outer surface cooling has been investigated experimentally. The results indicate that outer cooling of the containment surface is capable to efficiently reduce the pressure as well as remove the gas stratification inside containment atmosphere [8].

Containment spray systems are employed as one of the engineering safety measures to mitigate the consequences of severe accidents inside the containment in WCRs. The role of a spray system includes controlling containment thermal hydraulics by suppression of containment pressure and temperature and wash out of radioactive materials (aerosols, iodine). As illustration, the characteristics of a reactor typical spray nozzle are shown in Table 1.

TABLE 1. SPRAY CHARACTERISTICS OF A REACTOR TYPICAL SPRAY NOZZLE

CHARACTERISTICS	APR 1400, FRENCH PWR
Nozzle type and spray shape	Full cone
Spray angle	66°
Number of nozzles	506
Spray nozzle diameter	10 mm
Spray flow rate	1.11 kg/s
Droplet size (d_{32})	200–1000 µm
Droplet initial temperature	20°C, 60–100°C (recirculation mode)
Upstream pressure	3.5–5.25 bar (350–525 kPa)
Gas composition	Steam, hydrogen, air

Spray systems are shown to be effective in pressure reduction by cooling down the atmosphere and also to mix the containment gas atmosphere. In order to achieve a thermal equilibrium between spray droplets and surrounding gas atmosphere, a minimum falling distance is required [9].

The transient heating of the droplets as a function of the falling distance can be described as a first approach [9]:

$$T(z) = T_{atm} - (T_{atm} - T_0)e^{(-\frac{z}{w})} \tag{6}$$

with $T(z)$ being the droplet temperature after falling distance z, T_{atm} the temperature of the vessel atmosphere, and T_0 the initial droplet temperature. The parameter w, which can be determined experimentally, describes the overall heat transfer between atmosphere and droplets including the heat of condensation.

As spray induced steam condensation might also increase the volumetric hydrogen concentration leading to more severe combustion loads, the start time of spray operation is crucial. The containment spray system are generally designed to start automatically by an appropriate accident signal and also switch automatically from the fresh-water injection mode to the recirculation mode to ensure continuous operation until the design objectives of the spray system have been achieved.

PARs are an established measure of hydrogen mitigation in light LWRs containments. The enthalpy of reaction at the catalyst surface provides a driving force for buoyancy induced flow over suitably arranged catalyst elements, without mechanical assistance or outside power. The action is self-starting in response to the presence of hydrogen with available oxygen and continues until the hydrogen (or oxygen) in the enclosure has been completely consumed. The natural convective flow loop produced by PAR operation promotes mixing of the combustible gas mixture in the containment. As PAR does not need an active energy source, the availability of mitigation measures even during station black out scenarios can be ensured.

Natural convection is increased by the chimney effect of PAR housing, which in turn depends on temperature difference between PAR (T_{PAR}) catalyst and surrounding environment (T_{amb}), and PAR chimney height (h). The functional relation between the natural flow v_{nc} which is produced by the heat release of the catalytic cartridges and the chimney height h is given with:

$$v_{nc} = \sqrt{\frac{(2\,\Delta p)}{\rho}} \sim \sqrt{\frac{(2\,h\,g\,(T_{PAR} - T_{amb})}{\rho}} \qquad (7)$$

While taking into consideration PAR induced mixing of surrounding containment atmosphere, other parameters which might influence the mixing behaviour needs to be considered. As an example, an operating PAR subject to adverse flow conditions (e.g. counter current flow) may show different mixing behaviour.

2.3. HYDROGEN COMBUSTION

Hydrogen gas is colourless, odourless, tasteless and non-toxic gas. This means that hydrogen is not dangerous by itself. However, the main threat caused by hydrogen in the NPPs is its flammability. Hydrogen can chemically react with oxygen to generate water and thermal energy in a process called combustion. The heat released by hydrogen combustion is 120 kJ per 1 g of hydrogen:

$$2H_2 + O_2 \rightarrow 2H_2O + 120\ \text{kJ/g}(H_2)$$

Even though hydrogen is a flammable gas, it does not always burn when mixed with oxygen. There are necessary conditions for the combustion reaction to occur. The safety goal for hydrogen management is to understand these conditions specifically.

The flammability limits are usually determined experimentally. A mixture gas is flammable only when its relative concentrations of components are within a certain range. The threshold of this range is called the flammability domain. The minimum concentration of combustible gas within the range is referred to as ‘lower flammability limit’. The higher flammability limit refers to a maximum concentration of combustible gas within the range as the adequate amount of air is needed. The range of flammability depends not only on the hydrogen concentration, but also those of other species present in the gaseous mixture. The temperature, the pressure, the direction of propagation of the flame, and the mode of ignition also affect the ability of the mixture to burn. If the combustion is initiated where the composition of the gas mixture is not favourable to be sustained, the reaction rate is quickly reduced and the combustion is extinguished [1][2][10]. FIG. 4 shows an example of flammability limits for a hydrogen–air mixture as a function of temperature.

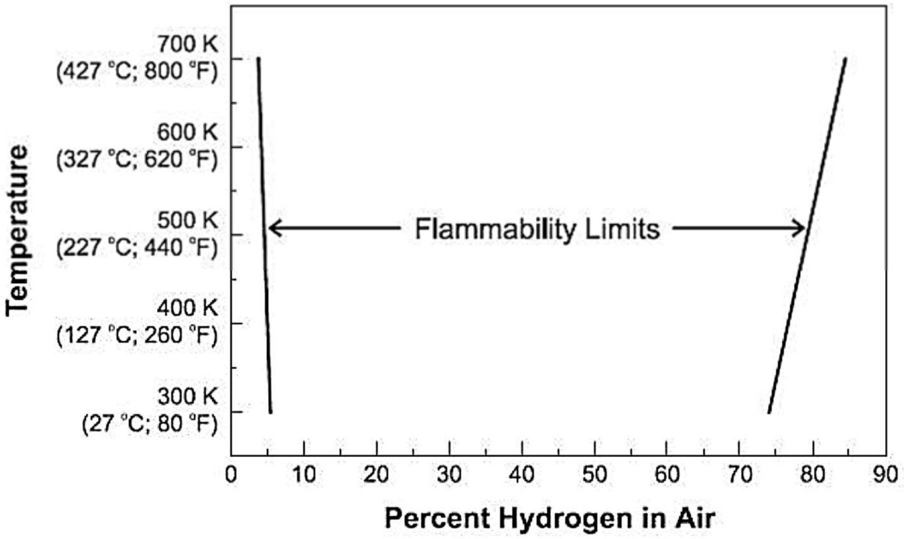

FIG. 4. Flammability limits of hydrogen–air mixture according to temperature [2].

In order for combustion to take place it has to be preceded by an initiating event, called ignition. Theoretically, the concentrations of hydrogen and oxygen may exceed the threshold without combustion if there is no ignition. For example, the reaction rate in ambient temperature is negligible; however, it can be accelerated by increasing the temperature above the ignition point or by presence of a catalyst such as platinum or in contact with hot surfaces.

2.4. HYDROGEN FLAME PROPAGATION

In case of combustion, hydrogen flame may either stay in a fixed position or propagate. The former phenomenon is called ‘standing diffusion flame combustion’ and occurs for the non-premixed mixtures of hydrogen and air. The latter phenomenon is called ‘flame propagation’ which can occur in a premixed condition. A flame is self-propagated by energy transfer from exothermic reaction to the unburned gas, which raises its temperature above the ignition point. This exothermic reaction is usually luminous.

FIG. 5 shows detailed configuration of a flame moving from right to left. The unburned gas is preheated by thermal conduction from the flame front where the combustion occurs. Then, its temperature increases due to temperature of ignition. At this temperature, the chemical reaction starts, and the combustion products are produced.

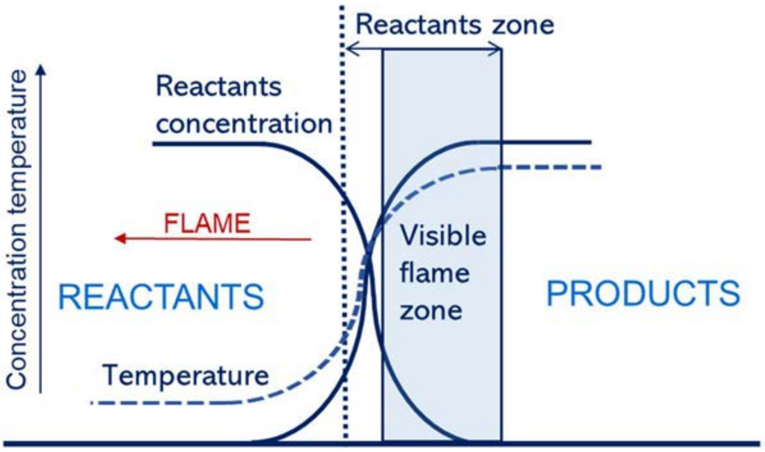

FIG. 5. Concentration and temperature profile in flame propagation, reproduced from [1].

The flame speed and the pressure induced by the combustion depend not only on the composition of a mixture but also on the momentum media, on confining geometry of the containment or building and physical obstacles within the combustion cloud such as piping and equipment [11].

Basically, there are two main flame propagation modes classified by the velocity of propagation:

- *Deflagration* occurs when the flame propagation is at subsonic velocity. It can be laminar deflagration, where velocity is relatively slow, or turbulent deflagration, where velocity is fast; and
- *Detonation* occurs when the flame propagates at supersonic velocity and is accompanied with the shock wave.

FIG. 6 shows the configuration of combustion according to different propagation modes [1].

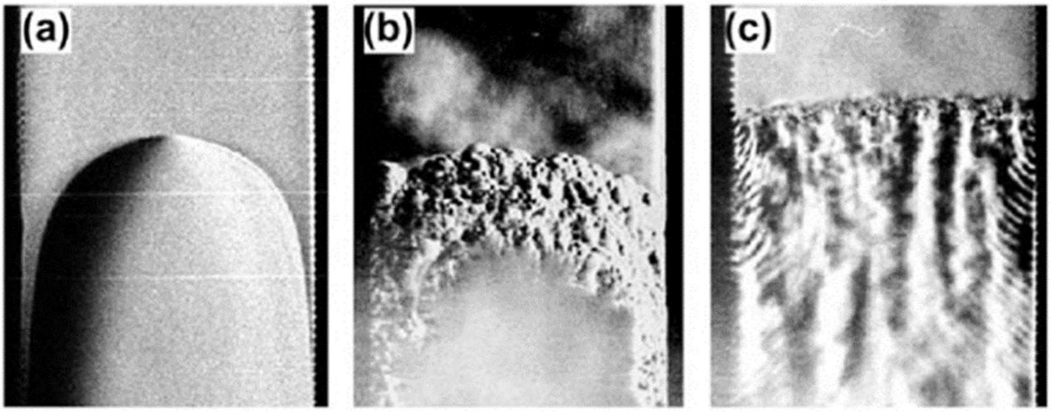

FIG. 6. Combustion regimes: (a) laminar deflagration; (b) turbulent deflagration; (c) detonation [1].

What is deflagration: When a mixture is ignited by a weak source, such as a small spark, the laminar flame starts and its propagation mechanism is a molecular diffusion of heat and mass.

The laminar deflagration propagates slowly, with a velocity of the order of 1–10 m/s. This flame speed depends on the chemical and thermodynamic properties of a gas mixture.

The laminar flame can accelerate and develop into a turbulent flame by increasing flow velocity and instability of flow ahead of the flame front. The flame speed of turbulent deflagration may reach hundreds of m/s. In turbulent deflagration, random fluctuations of the local flow are developed by large turbulent eddies as shown in FIG. 7. This wrinkling of flow increases the combustion rate and the flame velocity, and also intensifies the turbulence flow ahead of the flame. This strong positive feedback mechanism is established where the turbulent flame accelerates, high explosion pressure occurs and detonation can result in some cases [1][11].

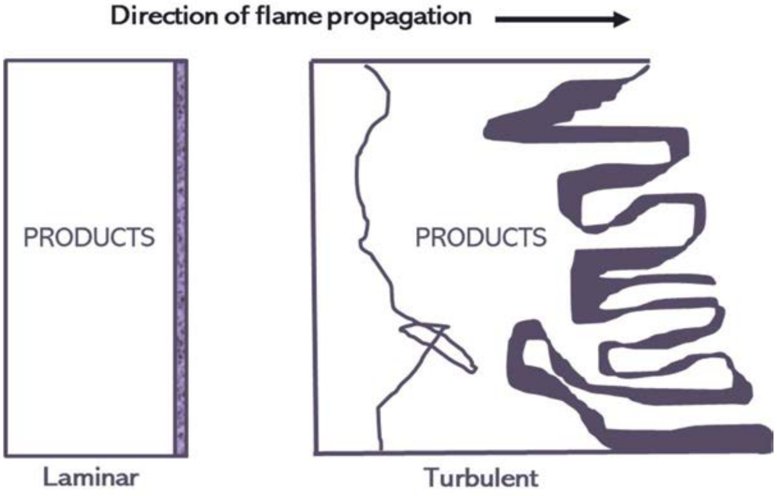

FIG. 7. Configuration of flame propagation for laminar and turbulent deflagration, reproduced from [11].

Highly intensified turbulence does not always enhance burning. The strong turbulence flame may lower the combustion rate due to rapid mixing of hot combustion products and cold unburned gases. If the temperature is lower than the minimum point where the combustion can be sustained, the flame will be extinguished locally, and the flame propagation will become slow. The quenching effect of turbulence is intensified as the unburned gas velocity increases. Therefore, the flame acceleration depends on both, the positive and negative effects of turbulence flows. This interaction between turbulence and combustion chemistry is highlighted in the Borghi diagram and shown in FIG. 8. The diagram is based on the following numbers:

— Damköhler number, *Da*, is defined as ratio of the integral turbulence time scale per chemical time scale, $Da = \frac{\tau_t}{\tau_c} = .\left(\frac{S_l\ l_t}{\delta_l\ u'}\right)$;

— Karlovitz number, *Ka*, is defined as ratio of the chemical time scale and Kolomogrov turbulent time scale, $Ka = \frac{\tau_c}{\tau_K} = \left(\frac{u'}{S_l}\right)^{3/2} . \left(\frac{l_t}{\delta_l}\right)^{-1/2}$; and

- Turbulent Reynolds number Re_t : $\mathrm{Re}_t = \left(\frac{u'}{S_l}\right).\left(\frac{l_t}{\delta_l}\right)$ where l_t stands for the turbulence integral scale turbulence, *u'* is the flow velocity fluctuation, S_l represents the laminar flame speed and δ_l the laminar flame thickness.

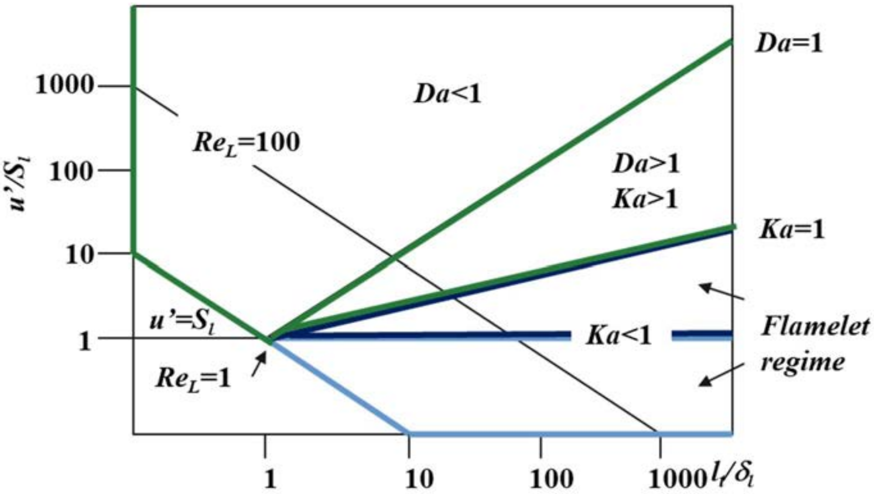

FIG. 8. Borghi diagram, reproduced from [12].

The three flame regimes can be distinguished:

1. Wrinkled flames and corrugated flames corresponding to Karlovitz number lower than 1 (Zone 1 and 2 in FIG. 8). In this case, the combustion reaction is faster than the turbulence Kolmogorov time scale. The flame front, where the combustion reaction takes place, is then thin and not affected by the turbulence. The turbulence will increase the flame front wrinkling and consequently the flame speed.
2. Distributed reaction zones where the chemical time scale is between the Kolmogorov and the integral time scales (*Ka* >1 and *Da* >1). In this case, the flame front is thicker. The turbulence can either increase the speed or lead to flame extinction.
3. Well stirred reactor where the chemical time scale is higher than the integral time scales (*Ka* >1 and *Da* >1). The turbulence can either increase the speed or lead to flame extinction. The turbulence can either increase the speed or lead to flame extinction.

Why is the deflagration to detonation transition (DDT) important: The DDT defines a transition phenomenon from the deflagration regime to the detonation regime of combustion, which is caused by shock waves. The turbulent flame accelerates to a point where pressure waves become shock waves which are faster than the speed of sound ahead of the flame front. If the turbulent deflagration accelerates, the shock waves are intensified. These shock waves compress unburned gas ahead of the flame front, increasing the temperature. The pre-compression of turbulent flows, in the period before DDT occurs, results in high velocity flame and overpressure. Overpressure is dependent on the degree of pre-compression in unburned gas ahead of flame front and may also be enhanced by the reflection of shock waves. Consequently, the combustion reaction is initiated by the pressure and temperature resulting from shock waves.

Multiple shock waves may develop and interact with each other moving in all directions. If strong shocks form sufficiently ahead of the intensified flame, hot spots appear in the unreacted gas. These hot spots lead to additional shock waves once the flame front reaches them, resulting in an unstable detonation which is called overdriven detonation. During the early stages of detonation, pressure and velocity peak in an overdriven detonation, and then decrease until a stable detonation condition is reached [13]. This is presented in FIG. 9 [14]. The detailed mechanism of DDT is still not fully understood. Currently, there is no exact theory to predict the condition where DDT occurs [15].

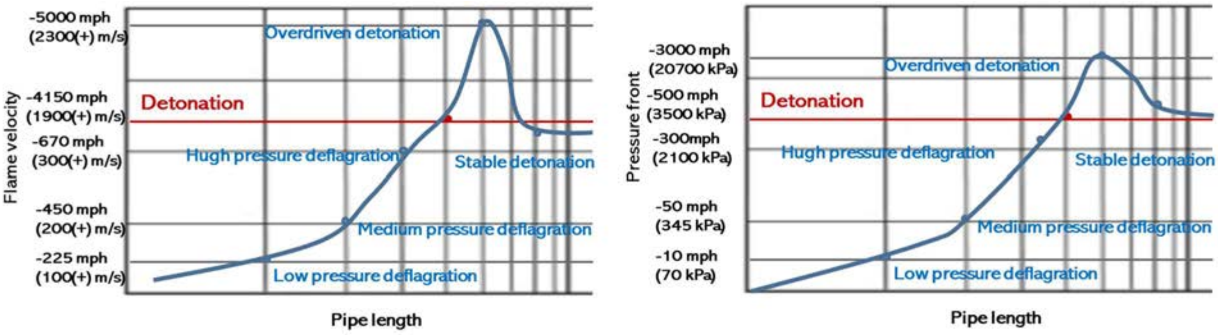

FIG. 9. Flame front velocity (left) and pressure during the DDT (right), reproduced from [14].

What is detonation: The detonation is a flame propagating at supersonic velocity, faster than the speed of sound. It is the most severe form of combustion. The flame speed of detonation can be 1,800–3,000 m/s. The peak pressure can rise by more than 200 bar (20 MPa) due to reflection and superposition of the shock waves. The chemical reactions are ignited in different ways between deflagration and detonation. In deflagration, the mechanism of combustion is heat and mass diffusion ahead of the energy release zone. In detonation, however, the combustion is ignited by strong pressure waves (i.e. shock waves), which compress unburned gas and raise its temperature to combustion.

Shock waves consist of curved shock segments as shown in FIG. 10. Detonation waves consist of three types of shock waves: Mach stem[2], incident shock and transverse wave [16]. The intersection of these three shock waves is called the triple point. The leading shock is separated into one strong shock, the Mach stem, and one weak shock, the incident shock. Combustion occurs closely behind the strong Mach stem, leading to a high post-shock temperature, whereas combustion is decoupled into weak incident shock. The reflected shock produces a transverse wave which propagates across the leading shock front. The Mach stem decays as it moves due to its curvature and the gradient field behind it. Then, the combustion front is decoupled from the Mach stem and the transverse wave propagates into the non-reacted mixture behind the incident shock. This will collide with another neighbouring transverse wave, generating a new Mach stem that is initially overdriven. Consequently, the Mach stem waves evolve into incident

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[2] Mach stem wave is a shock wave formed by the fusion of incident and reflected shock waves from an explosion.

shock and vice versa, resulting in the cyclical repetition of a fish scale pattern along the trajectories of the triple points in detonation [17].

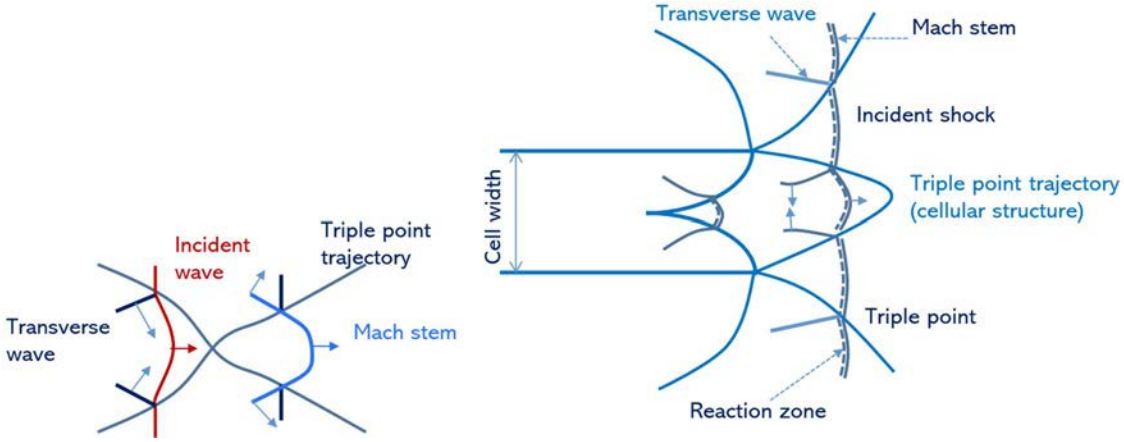

FIG. 10. Structure of detonation waves and triple point trajectory (left) and time evolution of fish scale detonation cell (right), reproduced from [16][18].

The detonation cell size or cell width can be used to evaluate the DDT. The mixture is more reactive and progresses faster for smaller cell sizes. Therefore, it is an important parameter for practical understanding and modelling of detonation. The cell size has been measured by experiments; however, there exist some variations as large as a factor of two in the reported studies. Hence, cell sizes should be considered as an approximate value to estimate the limiting conditions for DDT [1][11]. FIG. 11 is an example result of the detonation cell size according to hydrogen concentration of the gas mixture [18].

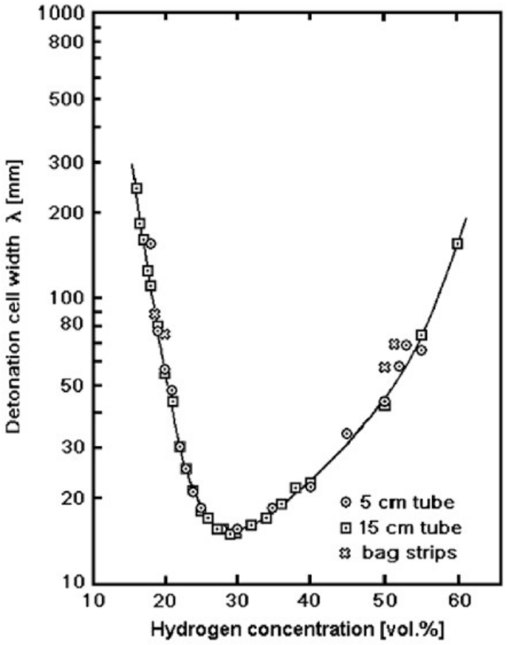

FIG. 11. Experimental measurement of detonation cell size for hydrogen–air mixture [11][18].

Another important parameter to assess the propensity of flammable mixtures to detonate is the run-up distance. The run-up distance is defined as the minimum flame propagation distance to transit to detonation. This run up distance depends on mixture properties (laminar flame, laminar flame thickness, and initial conditions) and the geometrical configuration where the flame propagation takes place. FIG. *12* presents the run-up distance for flame propagation in stoichiometric hydrogen-air mixtures in tubes with different diameters and various blockage ratio[3].

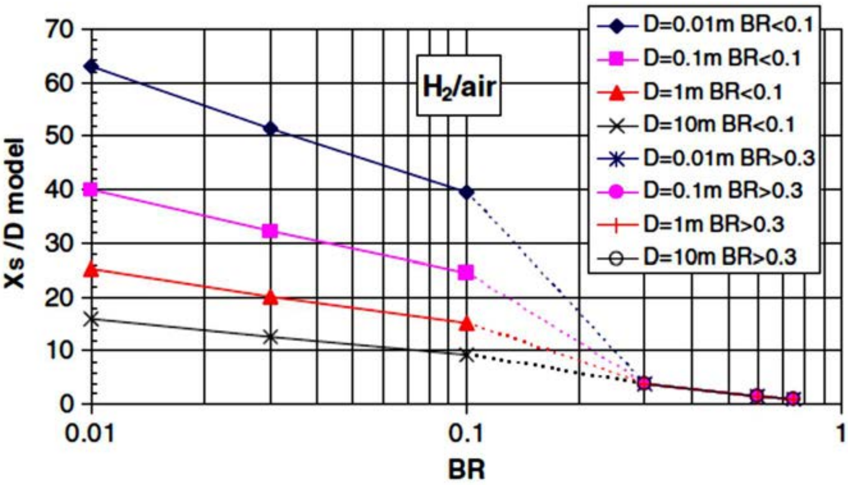

FIG. 12. Run-up distances in stoichiometric hydrogen–air mixtures at 298 K and 1 bar initial pressure as a function of blockage ratio (BR) for various tube diameters [19].

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[3] The blockage ratio BR, BR=1−(*d*/*D*), where *d* stands for the tube inner diameter taking into account the obstacle and *D* is the inner tube diameter without obstacle.

3. HYDROGEN BEHAVIOUR IN WATER COOLED REACTORS

This section provides the fundamentals of nuclear phenomena occurring within a reactor core of a water cooled reactor inducing a formation of hydrogen and its behaviour during the accident conditions. This includes the descriptions of in-vessel and ex-vessel hydrogen generation and conditions for its combustion, as well as associated risks. A good supplement to this publication is the IAEA–TCS–70 on Introduction to Water Cooled Reactor Theory with the Micro-Physics Simulator Lite Edition that provides fundamentals on nuclear reactor theory and water cooled reactor operation [20].

3.1. HYDROGEN GENERATION

There are several sources of hydrogen generation during the severe accidents in water cooled reactors. They can be classified as in-vessel sources and ex-vessel sources according to the place where phenomena occur:

- In-vessel sources:
  - Zirconium–steam reaction;
  - Steel–steam reaction;
  - Boron carbide–steam reaction.
- Ex-vessel sources:
  - Molten corium–concrete interaction;
  - Water radiolysis;
  - Corrosion of metallic materials;
  - Chemical reaction between corium debris and containment atmosphere;
  - Uranium–steam reaction.

Assessment of hydrogen risk and designing mitigation systems require accurate information about hydrogen source and generation conditions. Hydrogen generation rate strongly depends on boundary conditions such as masses of reactants, surface area, steam availability, surrounding temperature, and degree of pre-oxidization of materials. Hence, some models and numerical codes are available to simulate the progression of severe accidents, but the large uncertainties remain regarding hydrogen production. Consequently, further experimental research and model development are on their ways worldwide.

3.1.1. In-vessel source

In-vessel hydrogen generation is divided into early phase and late phase. In the early phase, core uncovering starts and metallic materials such as cladding and absorber, with relatively low melting points, melt and are relocated. The geometry of fuel remains almost intact. In the late phase, the UO_2 fuel melts and dissolves with other metallic materials, forming a ceramic molten mix. As the molten core relocates to a relatively cold area, a crust forms at its surface and causes channel blockage. The crust is continually eroded and reformed as the molten mix relocates to the lower vessel plenum, while particulate debris may be generated during this time [21].

Zirconium–steam reaction: During severe accidents, coolant may be depleted by decay heat of fuel resulting in core uncovering. This causes the temperature of fuel to increase as the amount of heat transferred from the uncovered core to steam is much smaller than the decay heat. The high temperature facilitates steam oxidation of cladding material, zirconium (Zr), and hydrogen to generate the heat. The amount of hydrogen produced by this oxidation is 0.0442 kg per 1 kg of Zr as follows:

$$\mathrm{Zr} + 2\mathrm{H_2O} \rightarrow \mathrm{ZrO_2} + 2\mathrm{H_2} + 586.6\ \mathrm{kJ/\ kg\ (Zr)}$$

Zirconium oxidation by steam is rapid, with a rate of 0.1–5.0 kg/s and speed of as much as ten times higher than for other hydrogen sources. These other hydrogen sources could be neglected. Within the first few hours of an accident, about 150–200 kg of hydrogen may be generated in a typical PWR and 2–5 times more than this in a typical boiling water reactor (BWR) [1].

If Zr is assumed to be fully oxidized, the mass of hydrogen produced in 1,000 MWe PWR is about 1000–1100 kg. The amount of hydrogen produced in BWR is almost three times more than that in a PWR due to a larger mass of Zr. Table 2 presents the mass of hydrogen generated from the steam oxidation of Zr in PWR, WWER and BWR.

TABLE 2. H_2 MASS GENERATED FROM 100% Zr OXIDATION WITH STEAM [1]

	TYPICAL PWR	WWER-1000 (RUSSIAN FUEL)	WWER-1000 (WESTINGHOUSE FUEL)	TYPICAL BWR
Zr	~ 26,000 kg	~ 22,630 kg	~ 24,765 kg	~ 76,000 kg
H_2	~ 1,150 kg	~ 1,000 kg	~ 1,095 kg	~ 3,360 kg

The oxidation kinetics of a cladding material, including Zr, for the intact core geometry is generally described by parabolic correlations as follows:

$$k^2 = \delta t = \mathrm{A}e^{-\frac{Q}{RT}} \tag{8}$$

where:

k —mass of oxidized Zr per square metre (kg)

δ—kinetic constant under Arrhenius equation[4]

t—time (s)

Q—activation energy (J)

R—universal constant = 8.314 kJ/mol/K;

T—temperature (K)

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[4] Arrhenius equation gives the dependence of the rate constant of a chemical reaction on the absolute temperature, a pre-exponential factor and other constants of the reaction.

Table 3 summarizes the developed correlations for the kinetic constant, δ, based on the experimental data. The unit of δ is $g^2m^{-4}s^{-1}$. Accident analysis codes extrapolate Zr oxidation kinetic correlations, based on the experimental data, to calculate hydrogen production for particular accident scenario. However, there are some limits to Zr oxidation correlations, because results have only been obtained for intact geometry with specific atmospheric pressure conditions and require correct modelling of the oxygen diffusivity. Recently, most severe accident codes use the Urbanic–Heidrick correlation because its result tends to correspond with the result of integral tests such as PHEBUS [22].

There remain large uncertainties in predictions of the hydrogen generation rate using the correlations shown in Table 3, which are developed only for the intact clad geometry. However, the Zr oxidation is affected by other boundary conditions including masses of reactants, surface area, steam availability, surrounding temperature, and degree of pre-oxidization of materials which change as core degradation proceeds. The following factors should be considered regarding hydrogen production:

- *Timing of cladding failure and loss of core geometry*: The later the core relocation occurs, the longer the time of intact geometry is. Hydrogen is generated more efficiently for an intact core geometry because the reaction surface with steam is large compared with that for the relocated molten core.

TABLE 3. CORRELATIONS OF Zr OXIDATION WITH STEAM IN CLADDING GEOMETRY

REFERENCE	CORRELATION
Baker-Just [23]	$\delta = 4.059\, e^{-\frac{190200}{RT}}$ (1,273 K < T < 2,123 K)
Urbanic-Heidrick [22]	$\delta = 0.036\, e^{-\frac{139841}{RT}}$ (1,323 K < T < 1,853 K)
	$\delta = 0.108\, e^{-\frac{138095}{RT}}$ (1,853 K < T < 2,123 K)
Prater-Courtright [12]	$\delta = 0.3622\, e^{-\frac{167200}{RT}}$ (1,783 K < T < 2,673 K) Relative error = ± 35%
	$\delta = 32.94\, e^{-\frac{220000}{RT}}$ (1,573 K < T < 1,783 K) Relative error = ± 42%
Erbacker-Leistikow [23]	$\delta = 0.524\, e^{-\frac{174284}{RT}}$ (1,073 K < T < 1,783 K) Relative error = ± 42%
Cathcart-Pawel [25]	$\delta = 0.362\, e^{-\frac{167117}{RT}}$ (1,273 K < T < 1,573 K)

- *Oxidation of U–Zr–O mixture*: Ceramic materials are liquefied and a U–Zr–O mixture forms during the core degradation stage during an accident. Experts regard the U–Zr–O mixture as a main source of oxidation in this stage. In the SKODA-UJP, preliminary test results present that the U–Zr–O mixture oxidation kinetic correlation is linear, and the oxidation rate is faster than that for parabolic Zr oxidation kinetic correlation for an intact geometry. This means that the hydrogen generation from a U–Zr–O mixture oxidation is greater than that in Zr oxidation. At this point, the parabolic correlation is not recommended to be used following the fuel melting. Nevertheless, the amount of hydrogen generated by a U–Zr–O mixture is estimated to be lower than that generated by an intact geometry because the reactive surface decreases.

- *In-vessel reflooding*: If the reactor core is reflooded, a large amount of steam is generated, causing high Zr oxidation kinetics and a high hydrogen generation rate. These have been observed in the TMI-2 accident and in several integral tests. In other words, the mitigation measures to remove decay heat in the long term could increase the hydrogen risk in the short term. However, FZK QUENCH experiments show that the results could be quite different depending on the reflooding timing. When reflooding occurs in an intact core geometry, hydrogen generation rate does not peak. This is expected because of the small contribution of ZrO_2 cracking and shattering, and cladding hydriding, which lead to the oxidation of the contained Zr. In contrast, when a slightly degraded core is reflooded, the hydrogen generation rate peaks during reflooding. The reflooding accelerates the oxidation of metal-rich mixtures such as Zr–O and U–Zr–O.

Steel–steam reaction: Hydrogen generated from steel oxidation may be accountable for 10–15% of the total quantity of hydrogen produced. Steel oxidation kinetic correlations were developed for an intact geometry and are presented in Table 4. However, there is no correlation for the failure of core geometry, similarly to Zr oxidation.

TABLE 4. CORRELATIONS OF STAINLESS STEEL OXIDATION WITH STEAM IN CLADDING GEOMETRY [Stainless steel oxidation rate, w, is expressed in mg/cm^2]

REFERENCE	CORRELATION
White [25]	$w^2 = 2.4 \times 10^{12}\ t\ e^{-\frac{353000}{RT}}$ (1,100 K < T < 1,300 K)
Baker [26]	$w^2 = 3.0 \times 10^{7}\ t\ e^{-\frac{209000}{RT}}$ (1,100 K < T < 1,300 K)

Boron carbide–steam reaction: Boron carbide (B_4C) is used as a neutron absorber in BWRs, WWERs and some western PWR designs. The BWRs and WWERs use B_4C powder inside stainless steel cladding and the western PWRs use sintered pellets.

The B_4C masses used in the PWRs roughly range from 200 kg to 300 kg and those in the BWRs with the same power are about four times larger. Table 5 shows the masses of B_4C, Zr and UO_2 used in each reactor type: typical PWR, typical BWR, French PWR, and WWER-1000.

When steam comes into a contact with B_4C during core degradation, the oxidation reaction occurs rapidly to generate hydrogen as well as various carbon containing gases such as CO, CO_2 and CH_4 and heat. Also, superficial boron oxide layer forms and then vaporizes. These reactions are summarized as follows:

TABLE 5. MASSES OF B_4C, Zr AND UO_2 ACCORDING TO REACTOR TYPE [1]

Parameter	Typical PWR	FRENCH P4-P'4 PWR	FRENCH N4 PWR	WWER-1000 (Russian fuel)	WWER-1000 (Westinghouse fuel)	Typical BWR
Thermal power (MWth)	3,600	3,800	4,270	3,000	3,000	3,800
B_4C (kg)	No B_4C	~320	~340	~270	~200	~1,200
Zr (kg)	~26,000	~28,000	~30,000	~22,630	~24,765	~76,000
UO_2 (kg)	~10^5	~$1.15x10^5$	~$1.24x10^5$	~80,100	~91,750	~$1.55x10^5$

a) $B_4C+7H_2O \rightarrow 2B_2O_3+CO+7H_2+740$ kJ/mol

b) $B_4C+8H_2O \rightarrow 2B_2O_3+CO_2+8H_2+770$ kJ/mol

c) $B_4C+6H_2O \rightarrow B_2O_3+CH_4+4H_2+965$ kJ/mol

d) $B_2O_3+H_2O \rightarrow 2HBO_2+661$ kJ/mol

e) $B_2O_3+3H_2O \rightarrow 2H_3BO_3+79$ kJ/mol

f) $3B_2O_3+3H_2O \rightarrow 2(HBO_2)_3+85$ kJ/mol

g) $CH_4+H_2O \rightarrow CO+3H_2$

h) $CH_4+2H_2O \rightarrow CO_2+4H_2$

i) $CO+H_2O \rightarrow CO_2+H_2$

B_4C oxidation significantly contributes not only to hydrogen production per gram of material much more than Zr oxidation, but also to heat release in the core. This effect strongly depends on the mass of B_4C in a reactor. Hydrogen is produced at about 0.15–0.3 kg per 1 kg of oxidized B_4C. For PWRs and WWERs, the hydrogen quantity from B_4C oxidation is approximately 10% less than that from Zr oxidation. Nevertheless, the B_4C reaction cannot be neglected because it may at least locally contribute to the generation of energy, hydrogen and other gases. Furthermore, the mass of B_4C in BWRs is much higher than in PWRs and WWERs. Hence, its contribution to hydrogen risk is more significant in BWRs.

3.1.2. Ex-vessel source

Molten corium–concrete interaction: When the integrity of a reactor vessel fails and molten corium drops into the dry cavity, which consists of concrete, the molten corium–concrete interaction (MCCI) occurs. MCCI is a complicated phenomenon and more so when the molten core is stratified into an oxide layer and a metallic layer. When corium with very high temperature contacts with the cavity surface, exothermic chemical reactions occur between corium materials and gases generated from concrete deposition such as H_2O and CO_2 and concrete becomes ablated.

During an early phase of MCCI, significant sources of hydrogen production are Zr and Cr contained in the corium. These metallic materials are oxidized with H_2O and CO_2, generating H_2 and CO from the following reactions [22][27].

a) $Zr+2H_2O \rightarrow ZrO_2+ 2H_2+600$ kJ/mol

b) $Zr+2CO_2 \rightarrow ZrO_2+ 2CO+600$ kJ/mol

c) $2Cr+3H_2O \rightarrow Cr_2O_3+ 3H_2$

d) $2Cr+3CO_2 \rightarrow Cr_2O_3+ 3CO$

It is expected that all amounts of Zr and Cr are fully oxidized within the first hour after onset of MCCI because there are very large masses of CO_2 and H_2O. Hence, the main limiting parameter of hydrogen generation is the remaining quantity of Zr and Cr which depends on the degree of oxidation during the in-vessel phase.

After Zr and Cr are depleted, the oxidation of Fe leads to long term hydrogen generation for several days compared to Zr and Cr for an hour. The hydrogen generation rate from Fe is 2 mol/s from the following reactions [22][27]:

a) $Fe+H_2O \rightarrow FeO+ H_2$

b) $Fe+CO_2 \rightarrow FeO+ CO$

In addition to H_2, a large amount of CO is generated during MCCI. Therefore, the study of flammability risk needs to be performed not only for H_2 but also for a mixture of CO and H_2.

Water radiolysis: Water radiolysis takes place in both normal operation conditions and accident conditions. It involves water molecule decomposition by radiation, producing diverse radicals. Eventually, hydrogen and oxygen molecules are produced based on the stoichiometric ratio. There are uncertainties for the conditions of elevated temperatures and the presence of solutes as the phenomena are generally studied for pure water at room temperature. However, the hydrogen generation rate is estimated to be very low under accident conditions. Hydrogen may be generated on the order of some hundred kilograms in three months and it can be sufficiently mitigated by existing measures such as hydrogen recombiners.

Corrosion of metallic materials: Zinc and aluminium are major sources of corrosion reactions resulting in hydrogen generation. These reactions are dependent on pH value. It is evaluated that the corrosion could generate in the order of 100 kg of hydrogen in some hours, which remains significantly less than that produced by other sources.

3.2. HYDROGEN COMBUSTION

3.2.1. Hydrogen distribution

During the core degradation, hydrogen is produced at a rate of up to 2 kg/s with contributions from various sources. The amount of generated hydrogen in the containment is usually over 1000 kg within the first seven hours of accident progression. If hydrogen is distributed homogenously, there is a very low possibility of hydrogen explosion which would threaten the integrity of the containment. However, hydrogen distribution is non-uniform as various phenomena take place in a multi-component atmosphere. Hydrogen concentration can be higher locally than the limit of flammability, resulting in its combustion. There is also a potential risk that DDT develops. Therefore, it is important to predict the hydrogen distribution.

There is a combination of various physical mechanisms which cause mixing in the containment atmosphere and influence the spatial concentration of different gases. The basic mechanisms are as described as follows [1].

Gas flow by inertial force: The initial gas flow in the containment is driven by inertial force which is caused by injected steam though a primary pipe break. The inertial flow may be the steam flow itself or other surrounding gas flow entrained by a steam flow. That is, convection loops have already been established in the containment atmosphere at the stage when hydrogen enters the containment.

Buoyancy by inherent density of matter and heat transfer: Buoyancy is an upward force caused by the density difference of materials. The gases with lower density move upward by buoyancy. The density difference is caused by either inherent characteristics of materials or heat transfer. As hydrogen is intrinsically a light gas, it usually moves upward. The hydrogen concentration of the mixture is expected to be relatively high in the upper part of containment within a few hours.

Containment structures such as walls, platforms, piping and equipment may be hotter or colder than containment atmosphere. In this case, heat is transferred between the structures and the atmosphere. The gas density is changed by this heating or cooling, and buoyancy driven flows are formed.

Molecular diffusion: Molecules tend to move from a region of higher concentration to regions of lower concentration by random motion. This movement is molecular diffusion. Hydrogen diffuses from highly concentrated regions to low concentrated regions in the atmosphere.

Steam condensation: Steam is normally expected to condense at the relatively cold surface of the containment structure. Some experimental results have shown that steam condensation in the bulk of the atmosphere also occurs. However, the contribution of condensation in the bulk atmosphere is smaller than that on the wall surface [1]. When steam condenses, the gas density becomes heavier than the nearby gas, and downward flows form entraining the hydrogen from a higher location to a lower location. Also, the local steam concentration decreases, leading to steam diffusion and Stefan flow[5] which are induced by fluids displacement for the vacancy of removed steam. Stefan flow entrain the hydrogen towards the low steam concentration region. Steam condensation not only influences the hydrogen distribution but also increases the flammability due to reduction of steam, which is an inert gas.

The influence of these phenomena on hydrogen distribution is dependent on the accident circumstances, accident scenario, accident stage and the location inside the containment. For example, when a pipe breaks and steam is ejected through the break, the inertial force is dominant near the break. As time passes, the buoyant force becomes more dominant [1].

Hydrogen distribution caused by physical mechanisms rely on the containment layout, the location of hydrogen release, safety systems such as sprays and fan coolers, hydrogen mass flow rate and mixing level with other gases [28].

Effect of a containment layout: Hydrogen distribution is influenced by general layout of the containment. As shown in FIG. 13, a containment with a missile shield is a good geometry to facilitate the mixing process if there exists a driving energy and a connection between the inner part and the outer part sufficiently available. The hot convection flows move upward in the inner part and then move downward in the outer part due to the cooling by the containment wall. FIG. 14 shows a diagram of dead end rooms, where there is no sufficient connection between the inner part and the outer part of the containment. In the early phase, the hydrogen released from the source accumulates in dead end rooms. In the late phase, the release rate decreases, and steam condenses, which increases hydrogen concentration. Hence, hydrogen explosion could occur in some dead end rooms if there is no hydrogen mitigation measure in place. In the case of a BWR or WWER containment with a pressure suppression pool, the hydrogen accumulation over the pool and steam condensation over the pool are intensified resulting in the increased hydrogen risk.

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[5] The Stefan flow, occasionally called Stefan's flow, is a transport phenomenon concerning the movement of a chemical species by a flowing fluid (typically in the gas phase) that is induced to flow by the production or removal of the species at an interface. Any process that adds the species of interest to or removes it from the flowing fluid may cause the Stefan flow, but the most common processes include evaporation, condensation, chemical reaction, sublimation, ablation, adsorption, absorption, and desorption. It was named after the Austrian physicist, mathematician, and poet Josef Stefan for his early work on calculating evaporation rates. [Wikipedia]

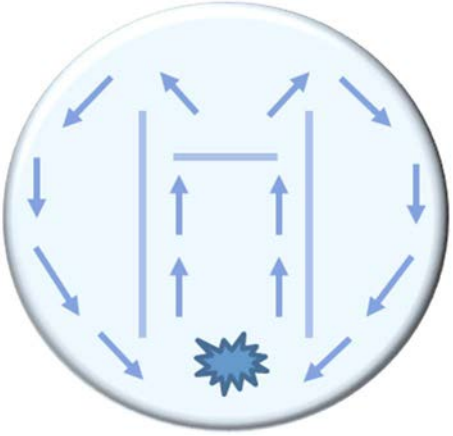


FIG. 13. General convection in the containment with missile shield, reproduced from [1].

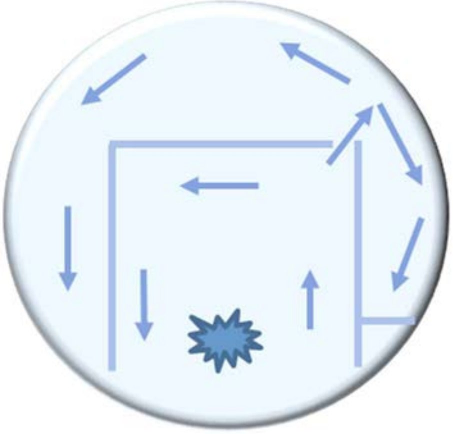
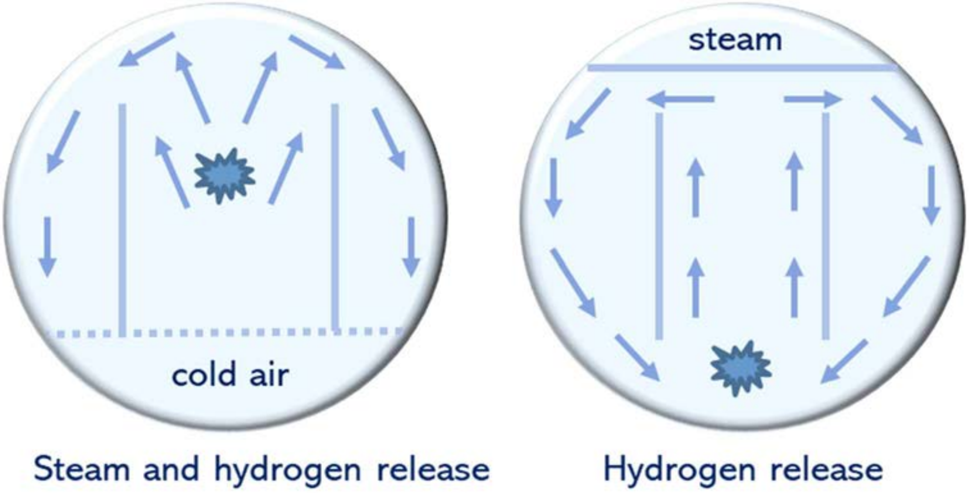


FIG. 14. Flows in the containment with dead end rooms, reproduced from [1].

Location of hydrogen release: The location of hydrogen release significantly affects its distribution. The hydrogen mixing experiments conducted in the framework of PHDR project have shown that the release of mass and heat in the lower part of spherical or cylindrical PWR containment makes good conditions for the mixing whereas high elevation releases tend to cause stratification [29]. In a PWR, in the case of a loss of coolant accident (LOCA), steam and hydrogen releases from the main coolant line or the surge line of the pressurizer. Inside the containment, the main coolant line is located at a lower position while the surge line is placed in a relatively middle or upper position. In case of non-LOCA, the release of hydrogen takes place in the coolant bleeding phase or on reactor vessel failure [1].

Safety systems: Safety systems such as sprays and fan coolers are used to decrease pressure inside the containment during an accident. These systems have two different effects regarding hydrogen distribution. They enhance the mixing process, preventing local formation of high concentration regions of hydrogen such as hydrogen pockets. However, they also contribute to steam condensation, which is the purpose of these systems. The inert gas is reduced and the relative hydrogen concentration increases resulting in the risk of combustion.

Release mode: The way hydrogen is released affects its distribution. A high mass flow rate could enhance convective motion in the containment atmosphere but also increase the hydrogen concentration, causing a risk for hydrogen combustion. Hydrogen distribution is also influenced by the combined release of hydrogen with other gases because of buoyancy. Hydrogen is usually expected to be released alongside steam. The steam dilutes the atmosphere as an inert gas and enhances mixing by buoyancy because of its low density. Figure 15 presents an example of mixing and stratification in the containment atmosphere according to hydrogen release mode, with or without a steam. Hydrogen is expected to be released together with steam in most accidents. FIG. 15(a) shows that both hydrogen and steam are released together in a relatively higher location (e.g. where the pressurizer relief tank is placed), while FIG. 15(b) presents the dry release of hydrogen without steam caused by a leak in the lower part. The containment is divided into inner and outer part by the missile shielding structure. In the former case, the light hydrogen–steam mixture moves upward, and the cold air is stratified at the bottom. In the latter case, the hydrogen is mixed with dry air, resulting in the higher density of mixture than that of the overlaying steam. Thus, the stratification between steam and air–hydrogen mixture takes place and steam cannot be penetrated.



(a) *(b)*

FIG. 15. Example of mixing and stratification according to release mode: (a) with steam and (b) without steam, reproduced from [1].

3.2.2. Hydrogen flammability

Two conditions should be satisfied for hydrogen combustion to occur in NPP containment during an accident. Firstly, the gas mixture of the atmosphere should be flammable. Secondly, the presence of an ignition source is required. Once combustion is initiated in an accident, the reaction may not be controllable until hydrogen inside the containment atmosphere entirely burns out. As an example, in the TMI-2 accident, hydrogen combustion took place by a random ignition source within the containment for approximately 12 s: less than 5% of hydrogen burned for the first 6 s, less than 40% of hydrogen burned for the next 3 s, and more than half of hydrogen burned in the last 3 s [1]. The pressure peak of about 0.2 MPa was observed by a deflagration but there was no detonation [1].

To determine whether the air–hydrogen–steam mixture in the containment is flammable, the Shapiro diagram is used for practical applications as shown in FIG. 16 [1]. The three sides of the diagram indicate the relative volumetric concentration of air, hydrogen and steam respectively, which are the dominant within the containment atmosphere during severe accidents. This diagram was empirically developed. The concentration of air and steam should be satisfied within a certain flammable range for combustion to take place. The steam reduces the flammability limit and dilutes the atmosphere as an inert gas. The outer curve presents the burn limit, within which a sustainable combustion could occur. A detonation can occur within the inner curve that is the detonation limit. According to Shapiro diagram, the gas mixture is an inert condition where the combustion cannot occur if the concentration of steam is greater than 55% [1][22].

These limits are not intrinsic properties of a mixture but dependent on geometry where they were obtained. However, this diagram gives useful information regarding the likelihood of hydrogen combustion and is widely used for different geometries. The limits also depend on the turbulence, the concentration of diluents, the temperature and pressure. However, the effect of pressure is not strong [1].

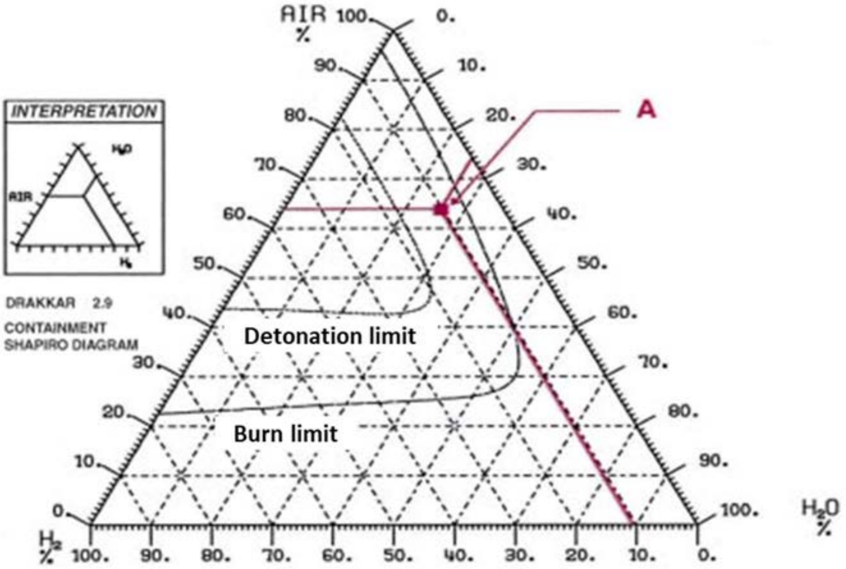


FIG. 16. Shapiro diagram for air–hydrogen–steam mixture [30].

During the severe accident late phases (e.g. MCCI), a large amount of carbon monoxide and carbon dioxide is generated and released inside the containment. The containment atmosphere will then contain flammable gas (hydrogen and carbon monoxide) and diluents (steam, carbon dioxide and nitrogen). In these configurations, the Shapiro diagram is represented as shown below in FIG. 17 [22].

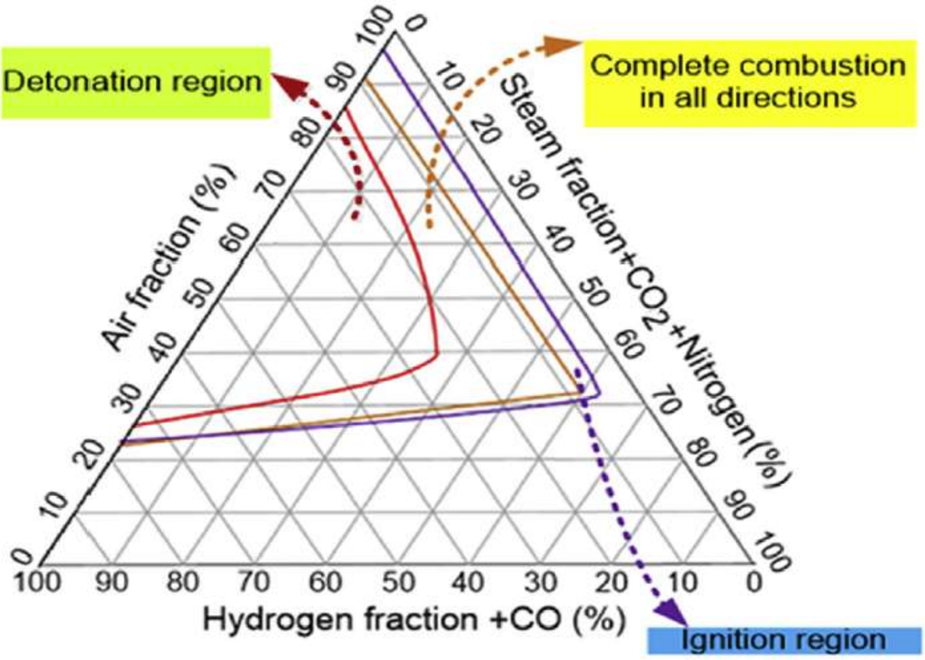


FIG. 17. Shapiro diagram for air–hydrogen–steam mixture with additional gases [22].

3.2.3. Ignition and combustion

What are the ignition modes: The ignition mode may have a direct influence on the type of flame regime. The ignition energy density can lead to a direct detonation if the hydrogen content is high. If the density is lower a deflagration regime occurs. Finally, if it is too low there is no combustion. The release of energy can be carried out via various means: electrical spark, jet of hot gas, hot surface, etc.

The mechanism governing the ignition is very complex and depends on the considered ignition mode. Thus, most of the available results are derived from various experiments. In the following paragraphs, a review of the minimum ignition energy of or the field of flammability of a combustible mixture air–hydrogen diluent is provided addressing the three ignition modes, by electric discharge (electric arc or spark), by a hot surface and by a jet of hot gases. These three modes are considered as plausible in the case of a possible combustion of hydrogen in the containment.

Ignition by electrical discharge: This mode corresponds to the contribution of the energy delivered through a spark, the electric arc. For most of the tests relating to the ignition of the hydrogen mixtures–air, the source of ignition is a flame or an electric arc. FIG. 18 shows the minimum energy needed for the mixture hydrogen–air to cause ignition, as a function of a concentration of hydrogen and a mixture pressure. This figure shows that the minimum ignition energy of a mixture air–hydrogen is extremely low: it is of the order of a few microjoules to a few millijoules. The U form curve shows a minimum of 17 µJ at atmospheric pressure for a stoichiometric mixture (29.53% of hydrogen). By its U shape, the minimum ignition energy is higher in the vicinity of the limits of ignition of the mixture: at atmospheric pressure; this energy is of approximately 600 µJ. It is therefore thirty times greater than the energy required to ignite a mixture with stoichiometric proportions.

Ignition by hot surface: This mode corresponds to an ignition by a hot surface. It is to define the minimum temperature capable of initiating a self-ignition of the combustible mixture. The extent of this autoignition temperature is not simple, because there is a period of self-ignition. This period of self-ignition is much shorter than the temperature of the gas. However, recent experimental data [32] shown in FIG. 19 indicate that the ignition thresholds increase from lean to rich mixtures, ranging between 1010 K and 1100 K with an additional increase to 1170 close to the upper flammability limit.

The PARs, under specific conditions, behave as ignition source. This ignition can be considered as an ignition by hot surface. This phenomena was highlighted experimentally in H2PAR [32] and KALI-H2 [34] programs. The recent OECD/THAI experiments [35] helped to confirm this phenomenon and to consolidate the ignition limit (i.e. hydrogen content leading to ignition) for different thermos-hydraulic conditions. These limits experimentally evaluated in OECD/THAI-2 project [35] for oxygen-lean atmosphere have subsequently been confirmed numerically [37] as shown in FIG. 18. This limit is extended to take into account the lack of oxygen that happens in late phase of severe accident. FIG. 19 shows the experimental results issued from the OECD/THAI2 project which were consolidated numerically using SPARK code [37].

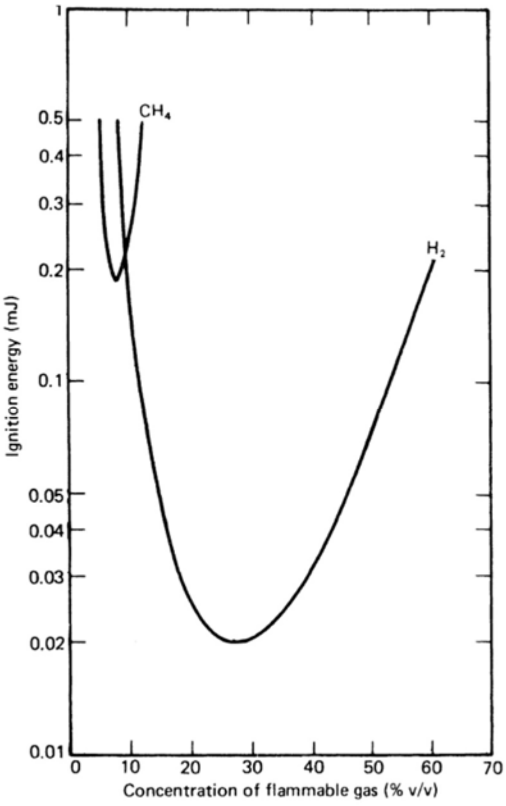


FIG. 18. Ignition energy of hydrogen–air mixtures compared to methane air mixture ignition energy [31].

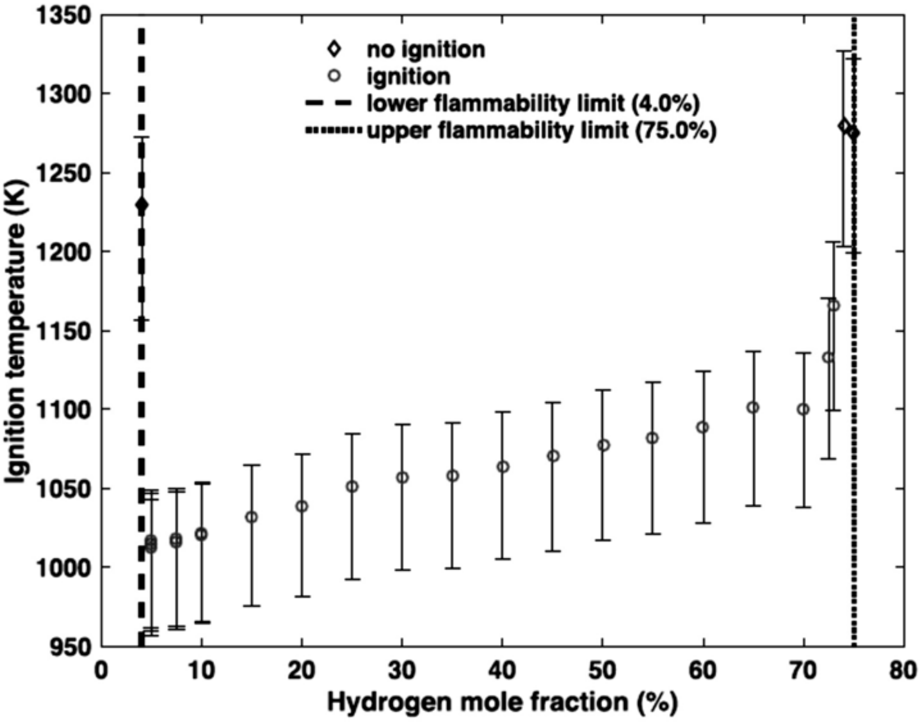


FIG. 19. Evolution of the ignition temperature threshold as a function of hydrogen mole fraction for hydrogen–air mixtures at 101 kPa T_o=300K; lower (LFL) and upper (UFL) flammability limits are shown as dashed and dotted vertical lines, respectively while the error bars indicate temperature uncertainty [32].

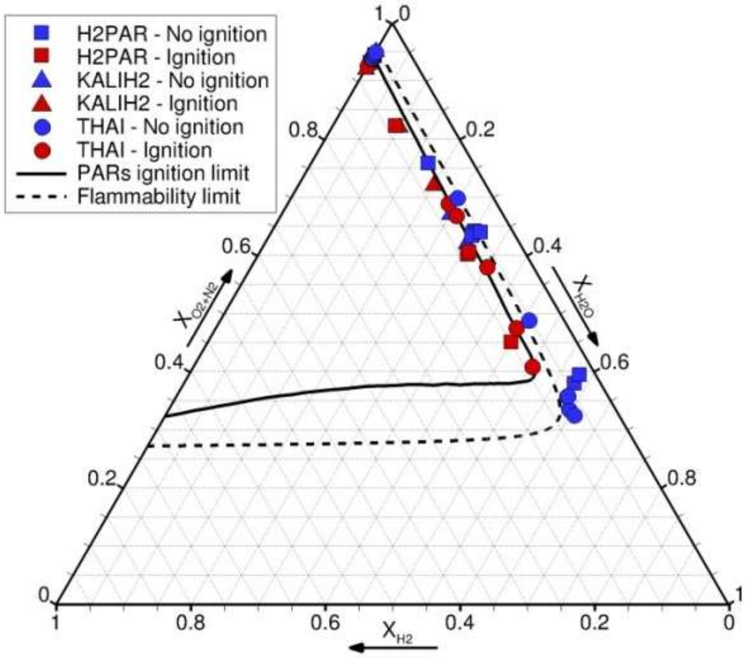


FIG. 20. Limit of ignition of hydrogen by the AREVA recombiners established with the code SPARK (at 1 atm) [37].

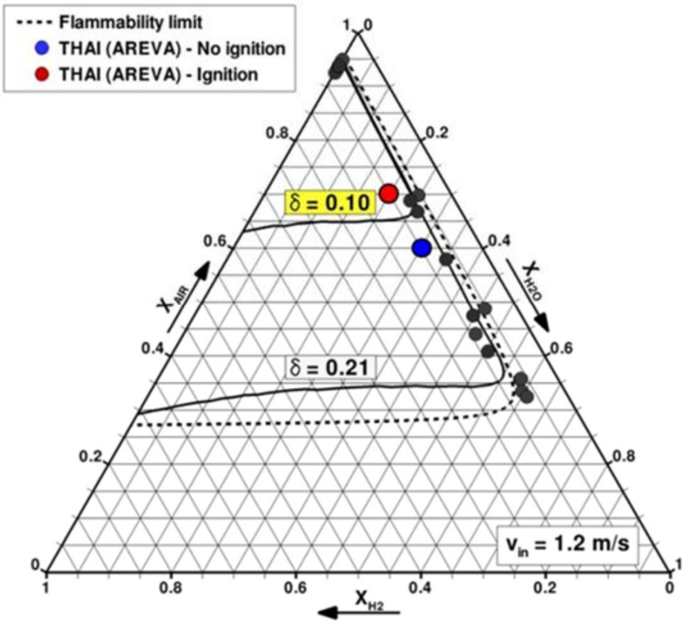


FIG. 21. Impact of lack of oxygen on the limit of ignition of hydrogen by the recombiners AREVA [35].

Ignition by hot gases jets: Hot jets can ignite flammable mixtures. In the case of severe accidents, the hot particles of corium ejected after the rupture of reactor vessel can constitute a hot jet which then can ignite the atmosphere in the containment and lead to a sharp increase of temperature and pressure inside the reactor containment (Direct Containment Heating).

To this end, this phenomena had been investigated experimentally in [32]; this study was to establish the limits of flammability of the mixture hydrogen–air–vapour hot jet at temperature around 2,700 K. The results show that the ignition by hot jets at temperature less than 1,500 K are similar to an ignition by hot surface, while the vapour concentration limiting ignition of a mixture depends on the initial temperature and pressure as shown in Table 6.

TABLE 6. VAPOUR CONCENTRATION LIMITS [31]

INITIAL TEMPERATURE (°C)	INITIAL PRESSURE (bar) (kPa)	CONCENTRATION LIMIT OF VAPOUR (vol.%)
100	1 (100)	39
100	1.5 (150)	41
130	1 (100)	48
130	1.5 (150)	51
130	2 (200)	54
130	3 (300)	62

Table 6 shows that the elevation of a temperature and/or pressure increases the vapour concentrations for limiting ignition and therefore broaden the field of flammability of a mixture. This dependence is similar to mode of ignition by spark.

What are combustion modes: All combustion modes (i.e. standing diffusion flames, laminar deflagration, turbulent deflagration and detonation) could potentially occur in a containment during severe accidents. If it is released into an oxygen–steam atmosphere and there exist ignition sources, hydrogen will burn as standing diffusion flame at the fixed release location. A typical example is in BWR suppression pools, where hydrogen is transferred from a drywell and the standing flames occur. These standing flames have thermal effects on the containment due to different heat transfer mechanisms: direct contact with the structure, radiation heat transfer in steam, and convection heat transfer between the gas and structure.

In hydrogen–air–steam mixtures and quiescent medium, hydrogen combustion, initiated within low ignition source energy, develops to slow deflagration. Pressure loads caused by this slow combustion are not severe, but thermal loads could threaten the safety components operability already installed inside the containment. The propagation aspect depends on hydrogen concentrations in a mixture. Thus, in the vicinity of the lower flammability limit, the flame propagates preferentially in the upward direction. This phenomenon is explained by the fact that H_2 is a very light combustible and the flame in this domain is very weak and not able to overcome the gravity. The buoyancy is favoring factor in the upward propagation. On the contrary, high initial hydrogen concentration lead to more energetic flame able to propagate in all directions as indicated in Table7. It presents flammability limits of hydrogen–air mixture for the propagation in different directions, which are obtained experimentally at ambient condition, pressure of 100 kPa and temperature of 25°C. The upward propagation occurs due to initial buoyant force when hydrogen concentration is between 4.1–6%. If hydrogen concentration is between 6–9%, combustion propagates in both, the upward and horizontal direction. For hydrogen concentration above 9%, combustion propagates in all directions with upward propagation being faster than downward propagation [1][22]. FIG. 22 shows flammability limits of a hydrogen–air mixture propagation for temperature range of 20–150°C, at pressure of 100 kPa and 250 kPa.

TABLE 7. HYDROGEN FLAMMABILITY LIMITS FOR VARIOUS PROPAGATION DIRECTIONS [1]

Direction of propagation	LOWER LIMIT (vol. %)	UPPER LIMIT (vol. %)
Upward	4.1	74
Horizontal	6.0	74
Downward	9.0	74

For hydrogen content between 4% and 9%, the maximum pressure due to the combustion is limited and well below the theoretical value that one would estimate based on complete adiabatic isochoric combustion FIG. 23. This is due to the fact that the flame could not propagate in the entire volume under quiescent conditions. Above 9% of H_2 and up to almost the upper flammability limit, the maximum pressure is very close to the calculated one which is again an indicator that the flame propagates in the containment. FIG. 23 illustrates this fact and compares the measured and calculated maximum combustion pressure in case of H_2/air mixtures initially at 1 bar (100 kPa) and 300 K.

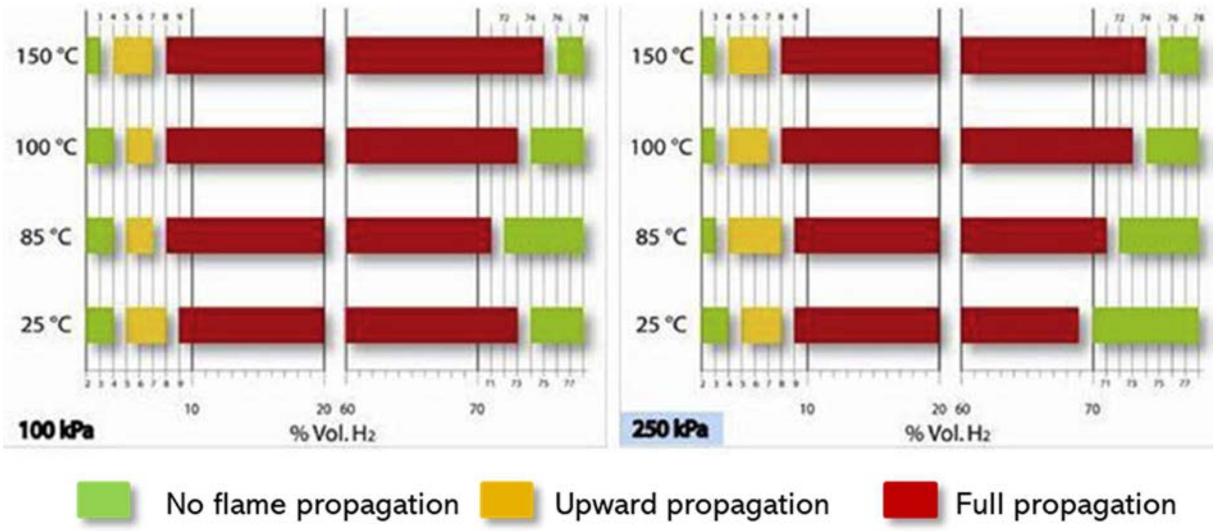


FIG. 22. Flammability limits for hydrogen–air mixture as a function of temperature and pressure[10].

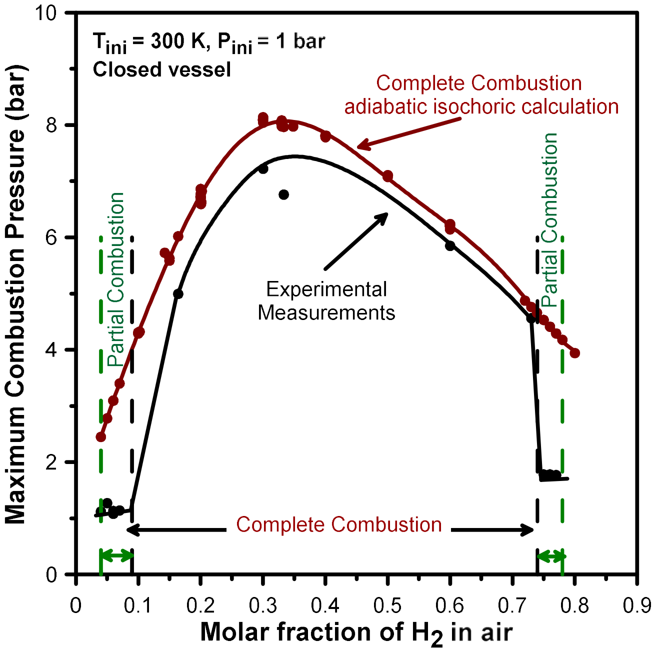


FIG. 23. Measured and calculated maximum combustion pressure of the hydrogen–air mixtures initially at 1 bar and 300 K (1bar = 100 kPa) [38].

For hydrogen concentrations above 8%, the combustion accelerates causing high pressure loads and becomes more complete. This flame acceleration is complicated and depends on not only hydrogen concentration gradient but also on hydrodynamic instabilities, the presence of diluent gases and geometrical factors such as degree of confinement and obstacle configurations. When concentration is higher than 10%, the acceleration intensifies more and its velocity may reach the speed of sound. The reaction may develop into a detonation in case of an extreme flame acceleration caused by turbulences. Detonation produces shock waves, resulting in high pressure loads. Pressure peaks and then decreases; a higher peak pressure does not necessarily cause higher loads to the containment structure. It is not enough to evaluate the structure vulnerability by using pressure peak alone, as the pressure declines very rapidly after it peaks [1].

Detonation could take place due to direct ignition or by transition from deflagration to detonation. However, the former event rarely happens in reactor accidents because it needs large amount of energy for a stable detonation. If hydrogen concentration is below 10%, the minimum required energy for a stable detonation is 100 kJ. If hydrogen concentration is above 18%, an ignition energy of 4 kJ is needed. Consequently, the deflagration–to–detonation transition is the only significant mechanism of detonation which occurs in the containment during a severe accident [1][22].

3.2.4. Flame acceleration and deflagration to detonation transition

After ignition, the flame reaches between 10 m/s and 30 m/s in a very short time. It will further accelerate due to the physical obstacles that are in its trajectory (such as but not limited to structures and equipment) and may transit to detonation. The pressure loads generated by these dynamic phenomena are so destructive that some structures inside the containment can be destroyed and the integrity of containment can fail. Therefore, a characterization of FA and DDT is very important to assess nuclear reactor safety.

There are two criteria which are used to evaluate the expected mode of hydrogen combustion:

- Criterion for flame acceleration; and
- Criterion for deflagration to detonation transition.

These are the empirical criteria derived from experimental research where geometrical conditions are tubes or tube–like arrangements. They are generally used to assess possible combustion modes during the nuclear accidents. However, discussion is still ongoing on whether these criteria can be applied to practical containment conditions (i.e. non-confined geometry with hydrogen concentration gradients). The validation of these criteria is still needed, and further development of criteria is expected.

What is a criterion for flame acceleration: A sigma criterion (σ) is an expansion factor of a mixture used to estimate the transition from slow deflagration to fast deflagration. It is defined as a ratio of reactant (unburned gas) density to product (burned gas) density at a constant pressure. This value is an intrinsic property of gas mixtures, depending on mixture composition and thermodynamic conditions such as temperature and pressure [39]. The necessary conditions for development towards fast combustion regime are as follows:

$$\sigma > 3.5\text{–}4 \qquad \text{for mixtures with } \beta(Le-1) > -2 \tag{9}$$

$$\sigma > \sigma^{*}(\beta) \qquad \text{for mixtures with } \beta(Le-1) < -2 \tag{10}$$

where $\beta(Le-1)$ presents the stability boundary of mixture flame with:

$\beta = E_a (T_b - T_u) / (RT_b^2)$ and $Le = \alpha / D$

where :

β–Zel'dovich (dimensionless) number named after Russian scientist Yakov Borisovich Zel'dovich provides a quantitative measure for the activation energy of a chemical reaction found in the Arrhenius exponent (jointly with David A. Frank-Kamenetskii, published a paper in 1938);

E_a–effective activation energy;

T_b–maximum flame temperature;

T_u–initial flame temperature;

Le–Lewis number is ratio of thermal diffusivity to molecular diffusivity:

If $\beta(Le-1) > -2$, the flame is unstable. For hydrogen–air–steam mixture that is a typical mixture in containment atmosphere, hydrogen rich mixture is characterized with $\beta(Le-1) > -2$.

If $\beta(Le-1) < -2$, the flame is stable. The hydrogen lean mixture is characterized with $\beta(Le-1) < -2$;

$\sigma^*(\beta)$–critical value indicating limiting boundary between two different combustion regimes, which is determined by β and Le. This critical value is obtained from the experiments for different geometries and scales. For rich mixtures, the sigma criterion is larger than critical value, in the range from 3.5 to 4, for the effective flame acceleration. For lean mixtures, the sigma value is larger than critical function value as shown in FIG. 24.

What is a criterion for DDT: The process of deflagration to detonation transition is divided into two main phases: a phase which creates the conditions for detonation (phase 1) and the onset of detonation (phase 2):

- *Phase 1*: There are several requirements for this phase to occur. The fast flame should be accelerated to the 'choked' or 'sonic' combustion regime where the flame speed is near isobaric sound speeds (about 500 m/s–1,000 m/s). The critical flame Mach number in the uncompressed gas mixture, which is the flame speed normalized to sound speed, should be over 1.5, and the Mach number of shock waves should exceed 1.2 so that the secondary ignition may be caused by their reflections off of obstacles and focusing surfaces [1].

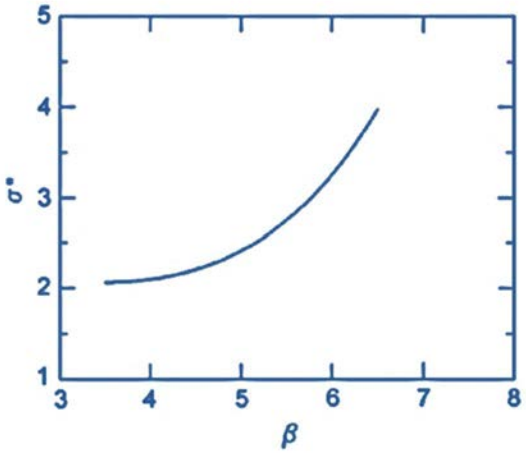


FIG. 24. Critical sigma σ(β) as a function of Zel'dovich number, reproduced from [1].*

— *Phase 2*: The characteristic geometrical size of the confined containment volume, L, should be larger than the chemical length scale which characterizes the mixture sensitivity for this phase. The L size should give a macroscopic measure of the gas mixture where the detonation initiates and develops. The length scale is usually regarded as the detonation cell size, λ, as presented in FIG. 25. The 7λ criterion has been proposed by Kurchatov Institute, Russian Research Centre as: $L>7\lambda$. This criterion has shown good agreement with experimental data for various geometry scales and mixture compositions [1][13]. However, several uncertainties should be considered when applying the 7λ criterion to reactor conditions. Firstly, the cell size data from experiments is limited to certain initial conditions and mixture compositions. In addition, the cell structure of a real detonation may be irregular, as the mixture can be non-uniform and have concentration gradients. As a result, it may not be fully applicable to apply the 7λ criterion in cases where complex mixture clouds may exist. Secondly, it is challenging to define the L size clearly for the complicated geometry of a containment. Thirdly, there have been no detonation experiments where the cell size was larger than 2 m. Hence, it is still questionable if cell size data can be extrapolated to values beyond 2 m and corresponding characteristic length exceeding 10–15 m [1].

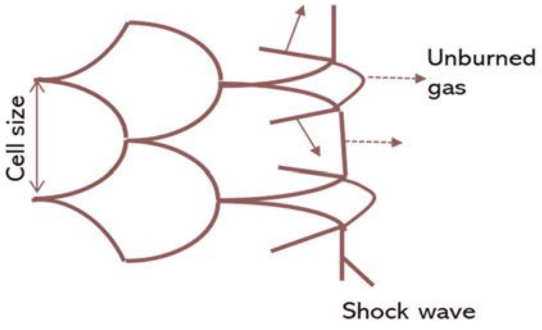


FIG. 25. Detonation cell size, reproduced from [1].

In case of accelerated flame or fast deflagration, flame speed is in the order of several 100 m/s. The resulting pressure for accelerated flame may exceed adiabatic isochoric complete combustion (AICC) pressure. The accelerated flames may impose heave pressure or differential pressure loads on internal structures or containment shell. The pressure loads impose by accelerated flames may also jeopardize containment structure. Flame acceleration may occur if the expansion rate, which represents the ratio of the gas mixture density before combustion to that after combustion, exceeds a threshold value (σ criterion). Additionally, the obstacles and vents can act as turbulence promotors. Turbulence increases the combustion rate, combustion completeness, and by this, pressure increase rate and peak pressure. As shown in the experiments, geometry plays an important role. Depending on containment geometry hydrogen flame acceleration can occur at hydrogen concentration as low as 9–10 vol.% with flame speed observed in the range of 200 m/s.

In the worst case the accelerated flame may also become a detonation (e.g. DDT). Detonations are combustion waves in which heating of the unburned gas is caused by compression from shock waves. Detonation flames (shock front) may speeds up to 2,000 m/s and peak pressure exceeds AICC pressure by far. Detonation waves travel supersonically and produce dynamic or impulsive loads on containment in addition to quasi-state state loads.

3.3. RISK OF HYDROGEN COMBUSTION

This Section summarizes the main aspects of hydrogen combustion in containment environments and provides brief review of experimental data of relevance to the scope of this publication.

Experimental analyses of flame propagation: Numerous experimental programmes have been conducted worldwide on flame propagation in a premixed atmosphere containing hydrogen [32][40][41]. These tests have two objectives: (1) to characterize the transition between slow and fast conditions and between deflagration and detonation; and (2) to produce a database for validating computational codes. There are two types of tests:

- Analytical tests to determine the laminar flame characteristics and to construct a database for qualifying the different flame conditions; and
- Dedicated tests for studying turbulent flames with the aim of validating computational codes and establishing criteria for characterizing possible flame regimes.

As was the case for hydrogen distribution in the containment, a state-of-the-art report on flame acceleration and deflagration to detonation transition was produced in 2000 by a group of international experts within the framework of OECD [32]. This report provides a description of the major experiments conducted in the facilities of BMC, NUPEC, VIEW, HTCF, FLAME, RUT, on flame acceleration and deflagration to detonation transition. Criteria for the transition between the different combustion conditions were developed based on the results obtained from the tests conducted in these facilities; these criteria were then refined as part of the European HYCOM programme and in the framework of the French ENACCEF programme.

Based on these program results and in order to identify the dangerous configurations that may lead to fast flame propagation and consequently to dynamic loads, prerequisite criteria, i.e. conditions required for the various combustion modes, have been developed. Two types of criteria have been defined:

- As described in Section 3.2.4, the criterion σ that concerns the flame acceleration; the value σ is the mixture expansion factor, i. e. the ratio between the cold gas and burnt gas densities at a constant pressure, and so is an intrinsic property of the mixture in question; the critical value σ^* above which flame acceleration is possible depends on the gases initial temperature and the flame stability and has been determined using the results of many experiments at different scales and in different geometries; and
- Similarly, the necessary conditions have been established for assessing the possibility of a DDT as described in Section 3.2.4; these are based on comparing a length typical of the studied chamber geometry with the size of the detonation cells (λ) characterizing the mixture sensitivity.

Within the mentioned criteria, it was easy to characterize gaseous mixture leading to fast combustion regimes as shown in FIG. 26. It can be observed that the transition from slow to fast flames occurs when the hydrogen concentration, in dry conditions, exceeds 10 vol.%.

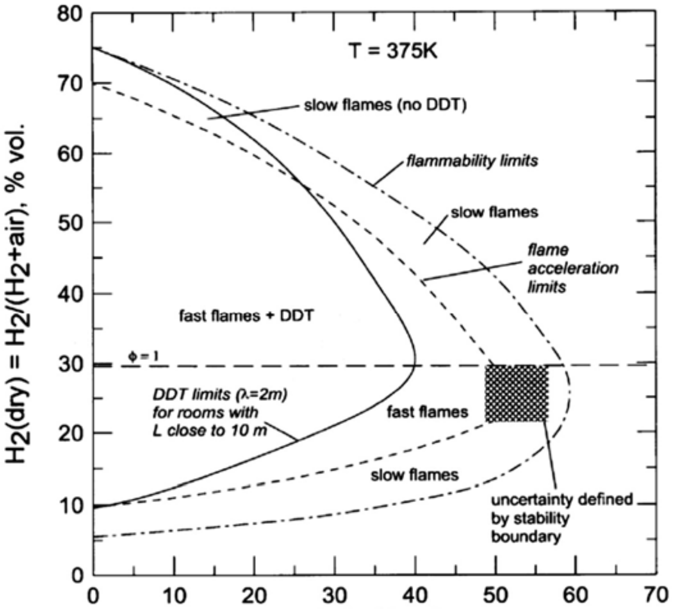


FIG. 26. Characterization of hydrogen–steam–air gaseous mixtures propensity to lead to flame propagation and flame acceleration [32].

Flame acceleration criterion extended to multicompartment geometry: The established flame acceleration (FA) criterion is extended to multicompartment geometry in framework of EU-HYCOM project (2000–2003) [42]. Originally, the criterion was developed based on tests in obstructed tubes with constant cross sections including cases of elevated temperatures and pressures and of steam dilution. The criterion gives a description of mixture properties that provide potential for effective FA in tubes with obstacles. Correlations for the critical expansion ratio σ are suggested in a following form:

$$\sigma > \sigma^*\left(\frac{E_a}{RT_u}\right) \tag{11}$$

where $E_a$is the effective activation energy, R is the gas constant and T_u the initial mixture temperature [42]. The effective activation energy for hydrogen–air–steam mixtures was assumed to be a function of equivalence ratio ϕ.This gives a correlation for the critical σ^* in the form of $\sigma^* = \sigma^*(T_u, \phi)$. The accuracy of the criterion was estimated as ±8% in critical σ^* value. For hydrogen–air mixtures at normal initial temperature and pressure, this gives the critical composition of 10.5 ± 1.3 vol.% of hydrogen.

In cases of complex flow geometry, combustion processes can be affected by the change of cross section along the flame path, or by lateral venting. Thus, the venting decreases the effective expansion of the products, and a more energetic mixture (larger σ) is necessary for strong FA. Thus, the effect of lateral venting on σ^* value is expressed as follows:

$$\sigma^* = \sigma_0^*(1 + 2.24\alpha) \tag{12}$$

where α stands for the ratio of the lateral surface opening per the cross section flame path. FIG. 27 shows the lateral venting effect on critical σ^* value based on experiments performed on RUT facility shown in FIG. 28 in the framework of HYCOM project.

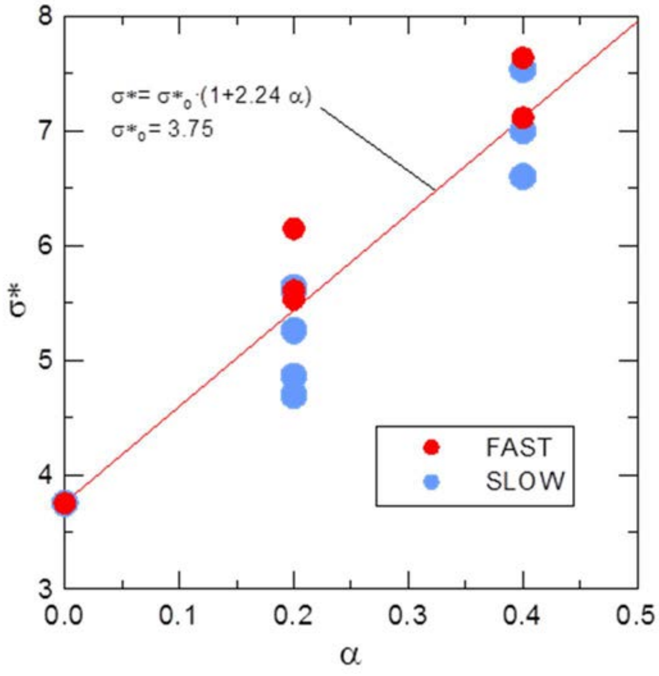


FIG. 27. Lateral venting effect on FA criterion [32].

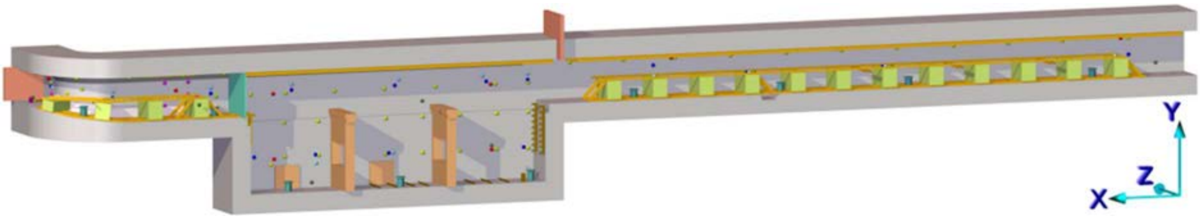


FIG. 28. RUT facility scheme (65 m length, height 6 m, canyon length 15 m, depth 2.5 m) [32].

In the opposite situation, reduction of cross section along the flame path can promote the FA. Thus, large flame area in the wide section of RUT generates an excess volume of hot products acting as a gas piston, which pushes the flame along the left channel as in the FIG. 29.

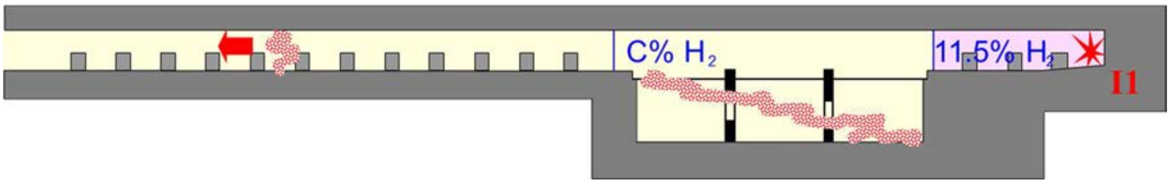


FIG. 29. RUT facility scheme and the piston effect on flame acceleration [43].

It was indeed observed that the flame accelerated strongly in this channel for tests with 10% hydrogen. High overpressures were generated in this case. The results show, however, that the decrease of hydrogen concentration down to 9% results in a very weak combustion process with low overpressures, even though the flame is pushed through the channel by the additional gas piston [42][43]–[44].

Concerning the uncertainty range of the σ criterion, the promoting effect of flow geometry on FA was found to be limited in strength, the existing uncertainty range of the σ criterion cannot be significantly reduced. This range covers the possible promoting effect of multicompartment geometry, which should be considered in reactor applications. It may be suggested that the application of the σ criterion should be combined with computational fluid dynamics (CFD) analysis, if further reduction of uncertainties is desired.

Flame acceleration criterion extended to heterogeneous initial mixtures: The sigma criterion was initially established for homogeneous gas mixtures and then extended to cover mixtures in which there are hydrogen concentration variations, on the basis of the ENACCEF program's results. The ENACCEF primary goal was to validate the σ criterion using tests conducted on a vertical structure representing a steam generator bunker opening up into the dome [45][46].

The ENACCEF facility as shown in FIG. 30 contains an acceleration tube forming the lower part of the containment and an adjustable dome forming the upper part of the containment. The acceleration tube, which was 168.3 mm in diameter and 3.3 m high, can be fitted with obstacles of various shapes, resulting in different blockage rates and notably an obstacle simulating an 11.12 litre steam generator. The adjustable dome's volume may be 780.9 litres or 957.8 litres. Several experimental campaigns were conducted considering stratified mixtures within upward and downward flame propagation. The performed experimental results led to the following conclusions [45]–[47].

- Hydrogen concentration near the ignition location have a great effect on flame acceleration;
- For negative gradients (ignition held in the hydrogen rich region of the mixture), the extension of the FA criterion was possible; and
- For positive gradients (ignition held in the hydrogen lean region of the mixture), the extension of the FA criterion was not possible.

Based on these conclusions and for the reactor applications, the time evolution of gaseous mixtures, inside the containment, satisfying the FA criterion is analysed. This analysis allows the definition of the ignition location and the ignition time that lead to high pressure load in case of combustion. Within, the combustion simulation results permit to deduce the expected maximal pressure loads.

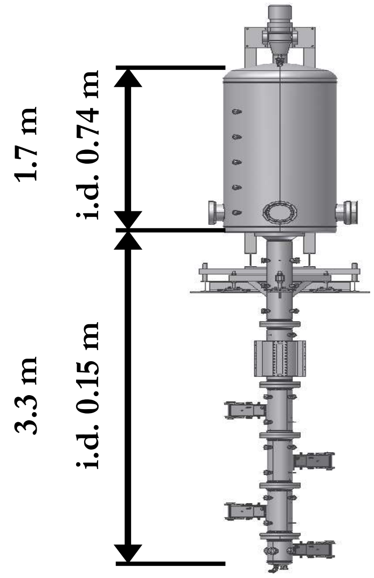


FIG.30. ENACCEF Facility [46].

Influence of other gases on hydrogen combustion: Inert gases, such as steam, N_2 or CO_2 are known to have inhibiting effects on hydrogen combustion behaviour. With respect to flammability limits, steam, N_2 and CO_2 show same characteristics [48]. The impact of steam and CO_2 on flame acceleration was also investigated experimentally in BMC test facility to show to be almost the same. No experiments with N_2 were performed to investigate its effect on flame acceleration in the BMC test facility [49].

In the late phase of a severe accident progression, the MCCI may take place. Depending on the type of concrete, substantial amounts of H_2, CO, and CO_2 can be released into the containment atmosphere and may form a flammable mixture with the residual oxygen (H_2 released during the early phase is assumed to be consumed either by a deflagration, by a standing flame or by PAR operation). Table 8 provides some examples of LFL calculated according to Le Chatelier's rule[6]: clearly the addition of CO lowers the H_2 concentration required for ignition at LFL. They also emphasize the influence of CO addition on combustion pressure, “which is significantly affected by the presence of CO in direct proportion to the contribution made to the energy content of the gas, i.e. while 5 vol.% CO in air will not burn, but when added to a 10 vol.% hydrogen–air mixture it definitely would, adding its energy to increase the pressure of the hydrogen burn. Thus, the addition of 5 vol.% CO to a 10 vol.% hydrogen–air mixture would increase the AICC pressure from 4.3 bar to 5.7 bar (for 1 bar initial pressure)” [50].

TABLE 8. FLAMMABILITY LIMITS OF H_2–CO MIXTURES [50]

H_2:CO RATIO IN FUEL MIXTURE	LEAN LIMIT (%)	H_2 IN MIXTURE (%)	CO IN MIXTURE (%)
1:3	8.16	2.04	6.12
1:1	6.06	3.03	6.03
3:1	4.82	3.61	1.20

Early stage of flame propagation (theory and experimental data of relevance to water cooled reactors): Once the mixture has been defined, the analysis of the likelihood of ignition and flame propagation regimes is analyzed. Firstly, the assumption that the flame will travel in a quiescent medium (no turbulence, no obstacles) is made. The speed at which the flame will propagate is important and is different from the laminar flame speed, $S_L°$, which is a characteristic of the combustible mixture composition and the thermodynamic state (initial temperature and pressure). The speed at which the flame propagates is named burning speed. The burning speed depends on the geometry as well. When a planar flame propagates inside a containment, the unstretched burning speed is proportional to $S_L°$, the proportionality factor in this case is the ratio between fresh and burnt gases densities, $\sigma = \rho_u/\rho_b$. However, in a real configuration a flame is never plane but curved and consequently it will be stretched. The stretch rate will also modify the burning speed and this modification will depend on the Markstein length[7], of these flames. Both the stretched burning speed and the Markstein length can be measured [51]. To take into account the stretch rate (or curvature of the flame), the following expression is used:

$$\left(\frac{S_b}{S_b^0}\right)^2 \ln\left(\left(\frac{S_b}{S_b^0}\right)^2\right) = -2L_b \frac{k}{S_b^0} \tag{13}$$

where:

S_b^0 – unstretched burning speed (m/s)

S_b – stretched burning speed (m/s)

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[6] *Le Chatelier's principle* is an observation about chemical equilibria of reactions: the changes in temperature, pressure, volume, or concentration of a system results in predictable and opposing changes in the system in order to achieve a new equilibrium state.

[7] *Markstein length*: the structure of pre-mixed flames in general flow fields is described using a multi-scale approach where one length scale, *δ*, characterizes the thermal thickness of the flame and the other, *L,* is a hydrodynamic length that characterizes the “size” of the flame. The *Markstein number* determines the effect of stretch on variations of local burning velocities [51].

k – stretched rate (1/s)
L_b – Markstein length (m)

The burning speed for lean hydrogen–air flames is plotted in FIG. 31 [52]. As the parameter σ increases strongly with the hydrogen content, the difference between $S_L°$ and $S_b°$ increases as well.

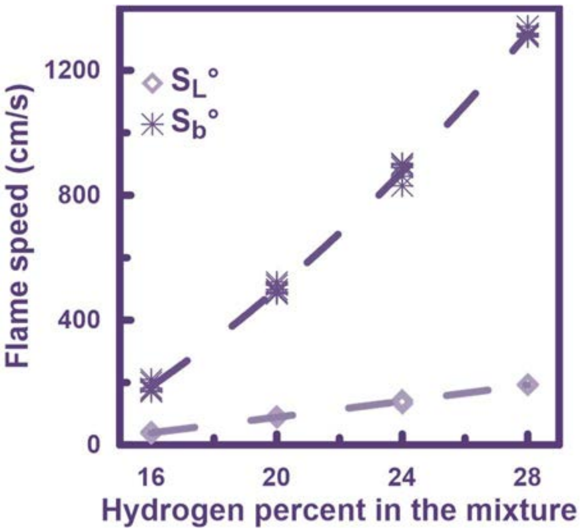

FIG. 31. Laminar flame speed, $S_L°$, and laminar burning speed, $S_b°$, versus the hydrogen molar percent for hydrogen–air mixture at an initial pressure and temperature of 1 bar and 295 K respectively, reproduced from [52].

The knowledge of S_b would be sufficient if the flame does not self-accelerate due to instabilities that can be of different nature (thermodiffusive, hydrodynamic, buoyancy, etc.). In a very recent work [52], the self-acceleration of the burning speed due to thermodiffusive instabilities is analyzed. The lean H_2–air mixtures are characterized by a Lewis number[8] lower than 1. These flames are inherently unstable, they develop a wrinkling of their surface as they grow in size. The onset of the flame folding occurs at a given radius of the flame, and this critical radius decreases when the hydrogen content is lowered. Figure 32 illustrates the evolution of the burning speed versus the stretch rate (which is inversely proportional to the flame radius). When the flame is smooth (before the onset of the flame folding), S_b decreases when the stretch rate decreases (as the flame radius increases). However, as soon as the flame begins to fold (marked by the symbol © in FIG.32), the burning speed increases drastically. The flame folding is responsible for a large increase in the flame area leading to a larger burning rate. One can then define a wrinkling amplification factor of the flame speed. This amplification factor depends not only on the hydrogen molar fraction in the mixture but also on the size of the flame. This factor varies from 1.2 (for the 28% H_2 case) to 1.8 (for the 16% H_2 case) in the investigated rate of flame radii (from 10 to 200 mm in diameter). As the flame accelerates in the containment, the flow ahead of the flame is set in motion and the interaction with the environment will lead

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[8] Lewis number (Le) is a dimensionless number defined as the ratio of thermal diffusivity to mass diffusivity. It is used to characterize fluid flows where there is simultaneous heat and mass transfer.

to turbulence generation. It is then important to study the effect of an initial turbulence on the enhancement of the burning speed.

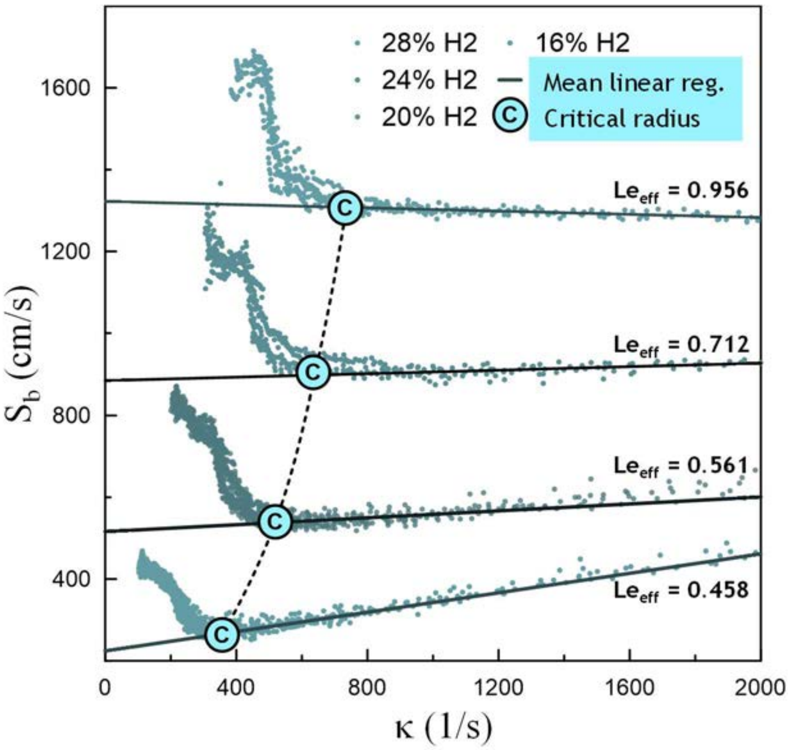


FIG. 32. Burning speed evolution versus stretch rate for hydrogen–air mixtures initially at 1 bar and 295 K, reproduced from [53].

The propagation of a spherical flame in a closed vessel equipped with several fans capable of generating a homogeneous and isotropic turbulence [53] was investigated. This study has shown that the increase of the flame speed depends on several parameters among them the size of the flame normalized by the turbulent integral length scale, the turbulence intensity normalized by the laminar flame speed and the Lewis number that accounts for the instable character of the flame:

$$\frac{S_T}{S_b^0} = (1.61 \pm 0.01)\left(\frac{r_{flame}}{L_T}\right)^{0.333\pm0.002}\left(\frac{u'}{S_L^0}\right)^{0.526\pm0.002} Le^{-0.140\pm0.005} \tag{14}$$

where:
r_{flame} – flame size (m)
Le – Lewis number
L_T – turbulent integral length scale (m)
u' – turbulent velocity fluctuations (m/s)
S_L^0 – laminar flame speed (m/s)
S_T – turbulent flame speed (m/s)

FIG. provides an illustration of the turbulent flame speed evolution versus the flame radius for various initial turbulent intensities ranging from 0.56 m/s to 2.81 cm/s. For a given initial turbulent intensity, the turbulent flame speed increase depends also on the size of the flame. At a radius of 60 mm, the flame speed is doubled when the turbulent intensity varies from 0 (laminar conditions) to 2.81 m/s.

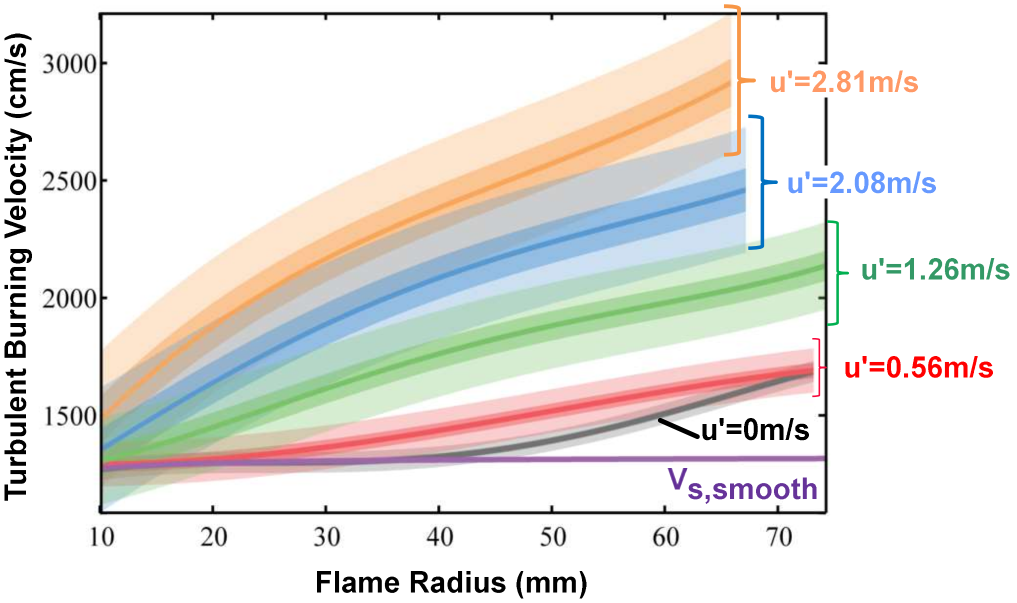


FIG. 33 Turbulent flame speed versus flame radius for different turbulent intensities; the mixture containing 28% of hydrogen in air was initially at 1 bar and 295 K [52].

Highly accelerated flames: Once the flame reaches between 10 and 30 m/s in a very short time (for a flame size of about 200 mm in diameter), it will further accelerate due to the physical obstacles that are in its trajectory (structures, equipment, etc.). According to [51][54], a simple criterion can be applied to discriminate *a priori* between (i) mixtures that have the potential to accelerate strongly, and hence induce a large pressure overload and (ii) mixtures that cannot sustain a string acceleration and as a consequence will induce a limited overpressure if not at all. This simple σ criterion which is estimated based on the thermodynamic properties (ratio between fresh and burnt gases densities, $\sigma = \rho_u/\rho_b$), the reactivity of the mixture (Zeldovich number, $\beta = E_a \cdot (T_b - T_u)/R \cdot T_b^2$ with E_a the global activation energy, T_b and T_u the flame temperature and the initial temperature respectively) and the transport properties of the mixture (Lewis number, Le), as described in this and in Section 3.2.4. This limit, defined as a critical value labeled σ^*, is calculated here using the following [51]:

$$\sigma^* = 0.09\beta(Le - 1) + 4.5 \qquad (15)$$

Equation (15) is based on the experiments performed in ENACCEF 1 [45][46]. The available experiments have supported the development and validation of combustion models. Nevertheless, deficiencies still exist that the scientific community is expected to address in order to reduce the uncertainty margins within the evaluation of the potential hazard in a given scenario. These deficiencies can be attributed to remaining uncertainties in the determination of the critical conditions, including critical values of mixture expansion ratio, in the detonation cell size data, the laminar burning velocity, the laminar flame thickness and the turbulent flame velocity. To overcome these limitations, a new programme has been launched in the framework of the French MITHYGENE project [55][56]. A new facility (ENACCEF 2) has been built dedicated to a detailed study of H_2–air–steam flame acceleration. This new facility is a vertical tube, about 8 m high and 230 mm i.d., equipped with several optical ports to detect the flame passage along the tube and large windows at specific locations along the tube to record the flame structure using high speed imaging techniques. These large windows will allow the use of high speed particle image velocimetry to measure the velocity field ahead of the flame induced by the latter.

Direct damage to the containment due to hydrogen combustion: Hydrogen combustion causes high pressure loads to a containment building. Containment may fail when exposed to loads about 1.5–2.0 times larger than the design load. Containment structures are equipped with penetrations such as hatches, pipes and cables. Failure mechanisms can be either a gross failure of a containment structure or failure of penetrations. If loads are applied, cracks are generated as an initial sign of failure. When cracks are sufficiently large, a gross containment failure may be prevented. The risk of containment failure can be estimated via a probabilistic assessment. FIG. 34 indicates the probabilistic curve of containment failure according to pressure. The higher the pressure is, the larger the failure probability.

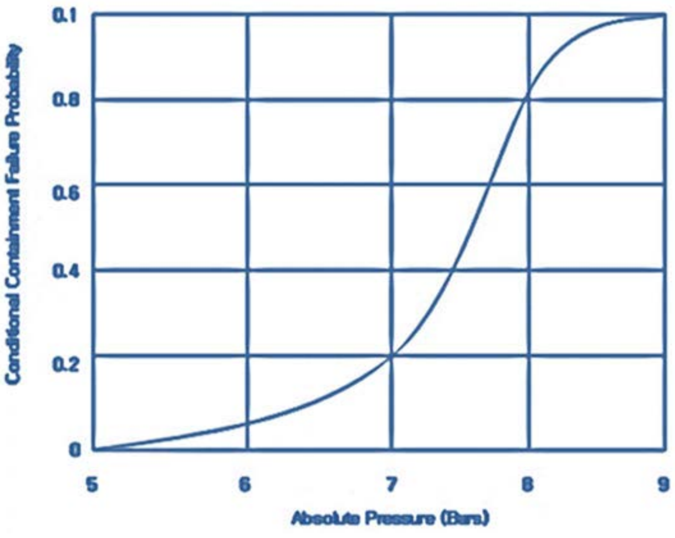


FIG. 34. Failure probability of containment according to pressure loads, reproduced from [1].

Another potential for damage is leaked hydrogen gas outside of a primary containment towards the secondary containment for the double containment concrete French PWR or the German KONVOI design, for example. Hydrogen gas can leak into the atmosphere of a secondary containment because it is impossible to perfectly seal a primary containment. In some cases, LOCAs or steam generator tube ruptures may lead to hydrogen gas bypassing primary containment, accumulating in the atmosphere of the secondary containment. As a result, risk of hydrogen combustion exists outside the primary containment as well as inside. Most primary containments are designed with a relatively low capability to withstand pressure loads from the outside whereas design margins against pressure loads from the inside are sufficiently large. Therefore, analysis and countermeasures for external pressure loads are necessary to enhance containment safety.

Indirect damage to the containment due to hydrogen combustion: In addition to direct damage, indirect damage may occur from hydrogen combustion. Containment consists of many compartments. If deflagration or detonation takes place locally inside the containment, some structures may be broken and detached, which can result in missiles crashing into the containment wall. The fact that hydrogen can be accumulated easily in some compartments increases the risk from indirect damage. Heavy missiles can threaten the integrity of the containment, especially in a steel containment. In general, concrete containment is more resilient than steel containment in absorbing missile impacts.

The walls of containment compartments are usually designed to bear the pressure differences of Design Basis Accidents (DBAs) such as a large break LOCA. However, it is difficult to analyse precisely what other loads the compartment walls could withstand as the composition of the walls are often not known in detail.

Mitigation measures promoting the convection of gas mixtures are helpful in preventing the severe local accumulation of hydrogen gas and decreasing the risk from resultant indirect damage. Fans or sprays are used to cause convective flow and compartment doors may be forced open by pneumatic devices to enable flow. It is important to notice that sprays can have negative results by condensing steam and making the mixture composition more favourable for combustion.

Secondary containment failure due to hydrogen combustion: Hydrogen which has escaped from primary containment can damage not only primary containment but also the secondary containment. Secondary containment is usually vulnerable to pressure loads from inside whereas it has been designed with sufficient margins for large external loads from earthquakes or airplane crashes. To alleviate hydrogen risk inside the secondary containment, filters can be used as a mitigation measure. The atmosphere of the secondary containment may be vented through filters to reduce the hydrogen concentration. Some of severe accident management guidelines, e.g. the Westinghouse Owners Group (WOG) severe accident management guidelines, addresses the issues of hydrogen accumulation and combustion inside the secondary containment.

4. MITIGATION MEASURES

4.1. HYDROGEN MEASUREMENT SYSTEM

Mitigation measures are often employed to avoid severe damage when the atmosphere of NPP containment is combustible during an accident. NPP personnel need information on the current status of the gas mixture inside the containment to select suitable and timely measures. Therefore, hydrogen concentration monitoring systems are important in supporting personnel to make appropriate decisions. There are two general methods for measuring hydrogen: measuring hydrogen concentration inside the containment or analysing sampled gas outside the containment.

4.1.1. Installation of measurement system inside the containment

A measurement method based on a catalytic reaction may be used inside the containment. The Beznau plant in Switzerland, the Doel units 3 and 4 in Belgium, and the Kozloduy plant in Bulgaria are using this system to measure hydrogen concentration [1]. Currently, many research projects are ongoing to develop an in-situ measurement system based on thermal conductivity detector or an electrochemical sensor which can endure the high temperature, pressure, irradiation and humidity which may be present during a severe accident [57]. In this context, IAEA released a TECDOC in 2017 on Assessment of Equipment Capability to Perform Reliably under Severe Accident Conditions [62]. The document covered aspects such as equipment capability assessment under severe accidents, equipment use in the scope of severe accident mitigation and monitoring, estimation of environmental parameters for instruments, demonstration of reliable performance under severe accident conditions.

4.1.2. Analyses of gas samples outside the containment

Generally, the sample extraction monitors that draw a gas sample through a sampling line are located outside containment, where the gas sample is analyzed and then returned to the containment. Germany has developed a new technique enabling sampling of gases inside the containment. Sampled gases are analysed using a mass spectrometer or thermal conductivity detector outside the containment as shown in FIG.35. These methods are accurate and have been used for a long period in many NPPs including nine PWRs and one BWR in Germany and the Kansai NPP in Japan, but there are several drawbacks [1][57]. The hydrogen can be diluted in the process of sampling gases from a containment. Also, the containment penetration is essential for installation of the sampling device, which increases the risk of containment leakage. The sampling system that is installed outside also needs to be protected to avoid any radiation exposure to personnel. On the positive note, the outside installed monitoring systems tend to offer higher system reliability as hydrogen monitors are installed outside containment and thus avoid a direct exposure to adverse conditions during a severe accident allowing long-term availability during a severe accident.

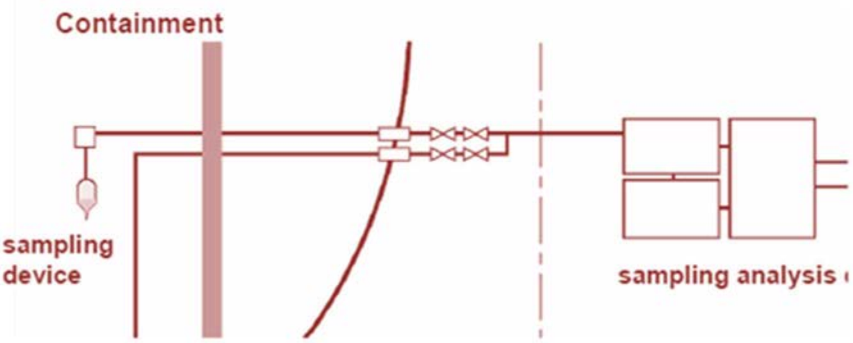


FIG. 35. Schematic diagram of sampling system to measure hydrogen concentration, reproduced from [1].

4.2. HYDROGEN MITIGATION SYSTEM

Mitigation systems are generally designed with an objective to mitigate the combustion threats posed by hydrogen by inducing high pressure and temperature loads which may jeopardize safety components or even containment integrity. Maintaining containment integrity is of fundamental importance to avoid fission product release (“source term”) to the environment as a comparison of the severe accidents in TMI-2, Chernobyl and Fukushima NPPs make obvious. There are many possible measures of mitigating hydrogen generation in NPP containments, which may vary depending on reactor design and the targeted severe accident management strategy.

Igniters: Hydrogen igniter systems remove hydrogen by deliberate combustion before dangerous amounts of hydrogen are accumulated. The combustion is a slow deflagration, as ignition occurs when the hydrogen concentration is relatively low. Hence, an increase of pressure and temperature as a result of combustion is not as high and pose only minor effects on the containment integrity.

The objective of the igniters is to minimize the consequences of potential combustion with an assumption that an ignition would eventually occur by a random source. The number of igniters, location of placement, and initiation time are to be determined appropriately for the effective control of hydrogen concentration. Igniters are to be installed at locations where the hydrogen release rate is expected to be high and where steam condensation is expected to occur. Some studies have indicated that as many as 150 igniters are needed to reliably maintain safe, low hydrogen condition [1]. When hydrogen concentration is lower than the flammability limit, the igniter system alone is not effective for controlling hydrogen concentration as the combustion does not occur.

There are three types of the igniters:

- *Glow plug igniters*: are electrical resistance heaters with hot surface temperatures of about 800–900°C. They are the most energetic ignition sources among the igniters. Ignition occurs when hydrogen concentration is very near its flammability limit. They can be operated manually (on and off), automatically (in response to LOCA signals) or semi-automatically (automatically but turned off by the operator). The disadvantage of glow plug igniters is the requirement of continuous and high power electricity supply from the outside, which is typically 150 to 200 W [1][58]. This means igniters may not operate in some severe accident scenarios such as a prolonged station blackout [1]. Cables and penetrations of a containment are also necessary to deliver the power.

- *Spark igniters*: Hydrogen requires the least energy for the spark ignition among combustible materials. Hence, spark igniters do not need relatively high power from the outside and are maintained by a battery power. FIG.36 shows the structure of a spark igniter. All components are enclosed in a water-jacketed metal housing which protects them from thermal and mechanical loads [1][58]. Spark igniters are actuated automatically by signals of pressure and temperature or hydrogen concentration during a severe accident. If the setpoint of actuation is low, unintended sparking may occur consuming the battery power. A high setpoint has the possibility of non-operation when needed. Therefore, the setpoint is optimized for battery depletion. The frequency of sparking can be determined in consideration of the time interval of flammable mixture formation. Sparking take place intermittently for the limited time until the battery power is totally depleted. Low frequency sparking is beneficial for battery conservation but may let substantial flammable mixtures form in the intervals. Combustion driven flows with velocity on the order of 10 m/s develop very rapidly following a spark ignition. As a result, gas composition near the igniter may change considerably in seconds. Spark igniters developed by Siemens (Germany), were designed to generate several sparks per minute for up to a week following an accident [58]. A remaining challenge is the compatibility of spark igniters with other electronics due to electromagnetic interference and spurious signals caused by spark discharges [1][58].

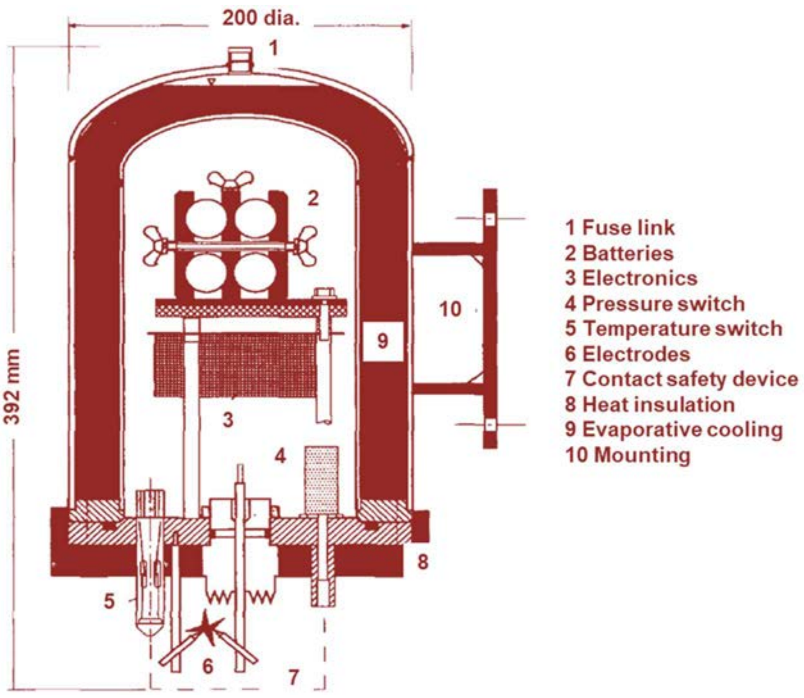


FIG. 36. Spark igniter, reproduced from [1].

- *Catalytic igniters*: initiate a combustion using a special catalyst. FIG. 37 shows a structure of catalytic igniter. A metal housing contains metal tubes coated with a catalytic material. Ignition wires are connected at the upper and lower ends of the tubes. Gas mixtures flow into the housing and the catalyst promotes the recombination of hydrogen and oxygen. This exothermic reaction generates heat to cause an ignition which then propagates to its surroundings [1]. Catalytic igniters are actuated automatically in response to the presence of hydrogen. The external power or battery is not needed. The disadvantage of catalytic igniters is their operation over a narrow range of mixture compositions compared with glow plug igniters and spark igniters, which means they actuate beyond absolute flammability limits. It may cause a problem in cases where hydrogen rich limit mixtures in which the margin between flammability limit and detonation limit is small. Their operation can also be limited by potential poisoning or fouling of catalytic surface. The response of catalytic igniters is not instantaneous as it takes time to react and produce heat. When catalytic igniters are used with catalytic recombination, both devices could fail by a common cause.

Comparison of these three types of igniters is summarized in Table 9.

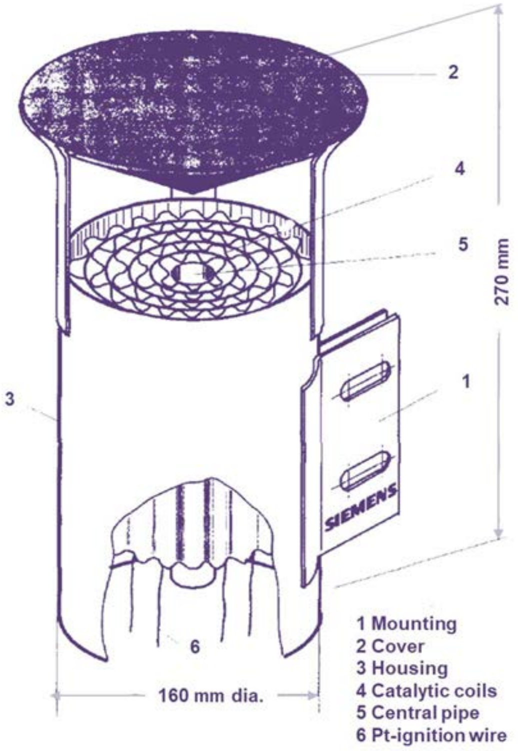


FIG. 37. Catalytic igniter, reproduced from [1].

Passive autocatalytic recombiners (PARs): consist of catalyst surfaces arranged in an open–ended enclosure as shown in FIG. 38. When hydrogen gas mixture enters the device at the bottom, hydrogen is recombined with oxygen, releasing heat at the catalyst surface [$2\mathrm{H}_2 + \mathrm{O}_2 \rightarrow 2\mathrm{H}_2\mathrm{O} + 120\,\mathrm{kJ/g(H_2)}$]. Then, natural convective flows are formed inside the enclosure, emitting the humid and warm air at the top and drawing-in fresh air at the bottom.

These natural convective flows promote the mixing of the surrounding atmosphere. PARs operate passively without any external power or operator's action [1][58]. PARs initiate when hydrogen concentration exceeds ~1–2 vol.% at the inlet [1].

TABLE 9. COMPARISON OF IGNITERS FOR HYDROGEN CONTROL IN NPP

TYPE	ADVANTAGES	DISADVANTAGES
Glow plug igniters	— ignite over widest range of compositions; — continuous availability; — robust; — operator controlled.	— rely on AC power; — high-power requirement; — containment penetration.
Spark igniters	— battery powered, do not rely on AC power; — easily back-fitted, no connections required.	— intermittent operation (in 5-s intervals); — not operator controlled; — weaker ignition source than for glow-plug igniters; — unavailable in long term; — electromagnetic interference from spark; — rely on triggering from LOCA signals.
Catalytic igniters	— self-powered, uses heat of H_2-O_2 reaction to produce ignition temperatures; — easily back-fitted, no connections required.	— operates over narrower range of compositions than do either spark or glow-plug igniters; — response to changing conditions not instantaneous; — potential for poisoning or fouling — rely on triggering from LOCA signals; — combined with recombiners, subject to common cause failure.

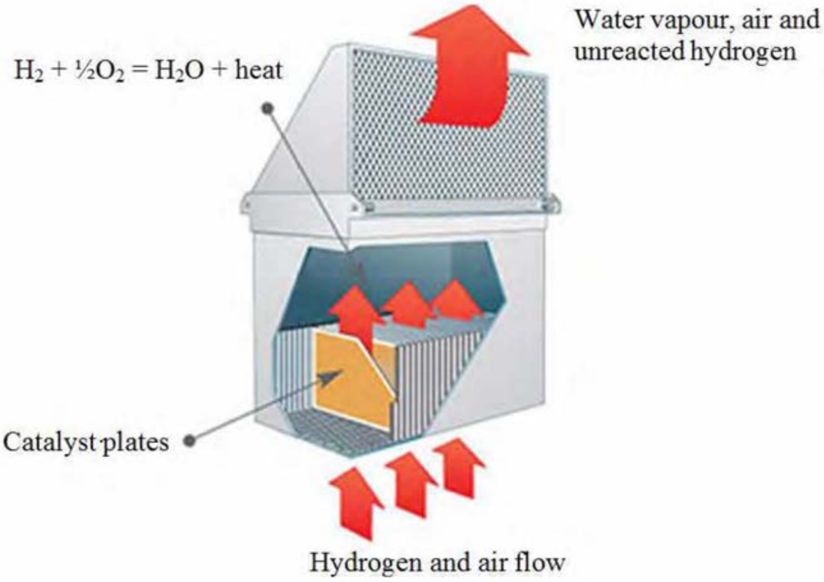


FIG.38. PAR components and flow of hydrogen [59].

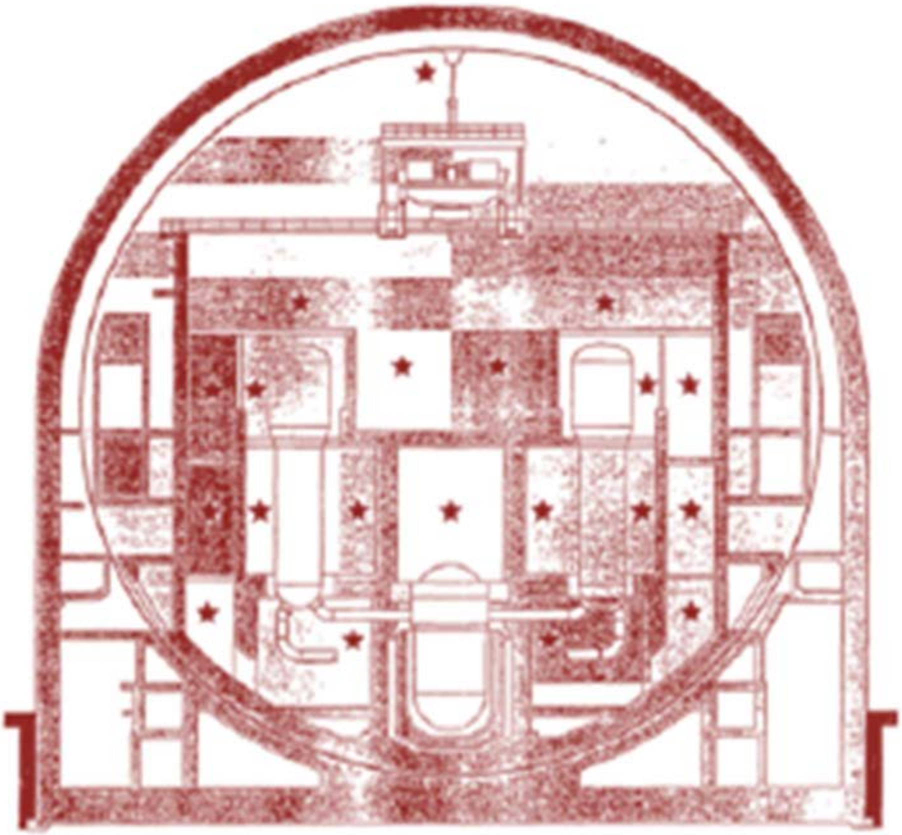


FIG. 39. Distribution of PARs in large dry PWR containment, reproduced from [1].

The capacity of PARs is limited by speed of natural convective flow, diffusion and mass transfer rate. Thus, they may not be optimal for mitigating high concentrations of hydrogen. Generally, 30–60 PARs are distributed in a typical PWR containment to deal with a wide range of hydrogen releases [1]. Figure 39 shows an example of PARs distribution in a large PWR, with stars indicating the PARs positions [1]. The arrangement of PARs is very important as it affects not only hydrogen concentration and flow patterns in the vicinity but also the PARs efficiency. Detailed code calculation need to be carried out to determine their optimal placement, considering factors such as hydrogen release location and rate, hydrogen distribution, geometry, operational constraints and other factors [1]. Significant progress has been made by the use of validated codes/models to design and assess hydrogen mitigation measures in NPPs and to take maximum benefit for accident management [60].

Overheated catalysts can cause unintended ignition if the exothermic reaction is intensified. In addition, a deflagration can initiate in the PARs in cases where hydrogen concentration higher than 6−8% (depending on steam content) due to the high temperature of the catalyst surface [35]. An example (AREVA PAR) of possible ignition area on ternary diagram by using minimum required H_2 / O_2 / steam concentrations is shown in FIG.40.

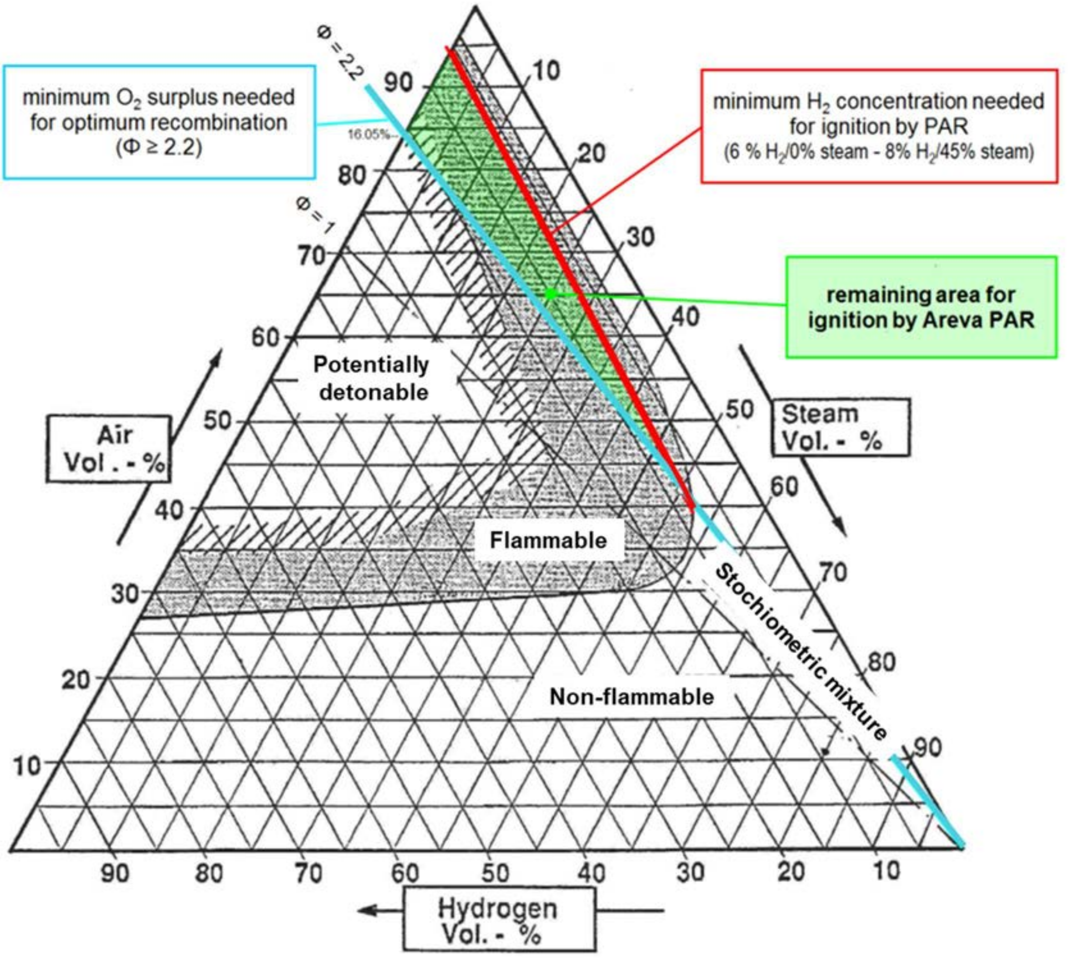


FIG.40. Ternary diagram: area of possible ignition by PAR, resulting from THAI tests [35].

Intertization: can be pre-intertization and post-intertization:

- *Pre–inertization*: dilutes oxygen by injecting a non-condensable inert gas such as nitrogen or carbon dioxide into part or all of the containment atmosphere during normal operation. Pre-inertization is used for most BWRs as the volume of containment is

relatively small. When oxygen concentration is below 5%, hydrogen combustion is impossible [1]. This method is applied in Japan, the United States and Sweden [1].
- *Post-inertization*: is used when an inertization is not practical during normal operation for large PWRs. It injects non-condensable inert gas into a containment atmosphere only after an accident starts. If the carbon dioxide or steam concentration exceeds 60% or nitrogen concentration exceeds 75%, the possibility of combustion is almost zero for any hydrogen concentration [1]. However, the large quantity of injected gas may cause a potential risk of containment pressurization. The plant operator decides the initial injection time, injection rate and injection position. These are decided in consideration of accident progression and its further evolution [1].

Venting system: intentionally vents containment, not only to control hydrogen concentration but also to depressurize the containment and remove heat during an accident. Fission products may also be filtered in the process to prevent significant release.

Containment pressure is relieved by venting the gas mixture, but steam may condense in long pipe lines or in filters and a dry gas mixture including hydrogen may appear in vent structures. Then, hydrogen combustion can easily occur in the vent systems, leading to piping damage or damage to filters and release of unfiltered radioactive materials. To prevent combustion near the vent system, pipe preheating has been suggested as a mitigation measure. The internal walls of pipes are preheated before opening the vent, and a saturation condition for steam in mixture gases is reached. However, long times are required until the temperature of internal walls reach the saturation condition. For example, the required time is about 20 hours to completely preheat the vent walls in the case of the U5 French containment. Sweden and Germany have applied another method in which filters are continuously heated. Electric power is needed for both measures, however this is generally not considered a problem as filters are expected to operate many days after an accident initiates with sufficient time to restore power even if power is lost in an early stage of an accident.

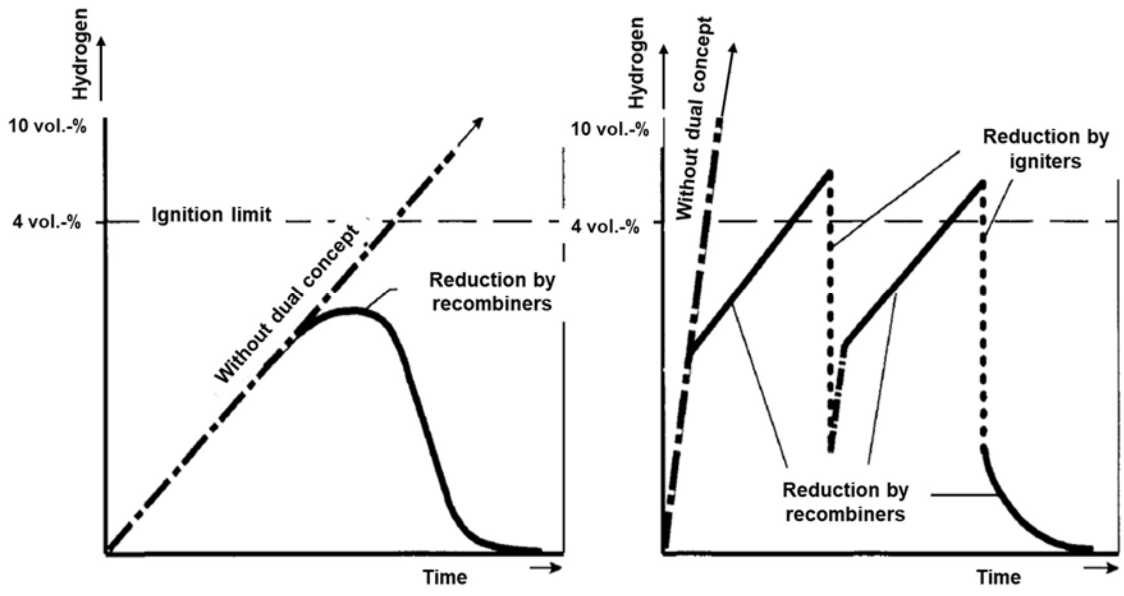
A recommended supplement publication to this topic is the IAEA-TECDOC-1812 on Severe Accident Mitigation through Improvements in Filtered Containment Vent Systems and Containment Cooling Strategies for Water Cooled Reactors:

> “One of the most important lessons from the accident at the Fukushima Daiichi nuclear power plant is that a reliable containment venting system can be crucial for effective accident management during severe accidents, especially for smaller volume containments in relation to the rated nuclear power. Containment venting can enhance the capability to maintain core cooling and containment integrity as well as reduce uncontrolled radioactive releases to the environment if the venting system has a filtration capacity. In general, a filtered containment vent system increases the flexibility of plant personnel in coping with unforeseen events. This publication provides the overview of the current status of related activities with

the goal to share information between Member States on actions, upgrades, and new technologies pertaining to containment cooling and venting."[61]

Strategic combination:

- *PARs and igniters*: Several PARs distributed inside containment deplete the hydrogen in case of low hydrogen release as shown in FIG. 41 (a). However, they alone may be insufficient to avoid flammable accidents for large releases of hydrogen due to their limited capacities. The combination of PARs and igniters, called 'dual concept', has been developed and tested in Germany [1]. The igniters remove a relatively large quantity of hydrogen by combusting gas mixtures when the recombiners are incapable of coping with high hydrogen concentration. Figure 41 (b) indicates the hydrogen concentration change with the dual concept of PARs and igniters is used for a case with large hydrogen release. This dual concept is used for the Loviisa NPP in Finland and the APR1400 in the Republic of Korea [1].
- *PARs and post-CO_2 injection*: Carbon dioxide is injected in amounts capable of preventing a DDT and a detonation during PARs operation. However, there remains the possibility of deflagration to occur. Also, the injection of incondensable gas produces additional pressure loads to the those from a deflagration. The amount of injected gas is limited to such an extent that the pressure loads by both gas injection and deflagration do not endanger the containment integrity [1].



(a) Low hydrogen release location *(b) High hydrogen release location*

FIG.41. Hydrogen concentration change with dual concept of recombiners and igniters, reproduced from [1].

5. INTERNATIONAL PROGRAMMES

International cooperation on hydrogen safety research has been a focal point to consolidate the knowledge as well as for development of common understanding on hydrogen safety issues in LWRs, e.g. state of the art reports, and code benchmarks as international standard problems (ISPs).

International experimental programmes of OECD/NEA and EC: The experimental and analytical efforts aimed at understanding hydrogen related phenomena or processes that may occur during an accident are required to support achieving a high level of confidence that nuclear reactor containment systems and components provide an acceptable level of safety. At international level, hydrogen safety related projects have already been running since long time, e.g. OECD/NEA SETH project with specific focus on containment thermal hydraulics was launched back in 2001. In SETH project, experiments were performed in PANDA test facility operated by PSI in Switzerland. The joint nuclear safety project OECD/NEA THAI was started in 2007 with an aim to investigate hydrogen and fission product behaviour in LWR containments under severe accident conditions. The experiments have been performed in the THAI test facility operated by Becker Technologies in Germany.

The Fukushima Daiichi NPP accident further affirmed the need to advance knowledge and therefore R&D on hydrogen propagation, conditions for explosion and safety of containment building and components. One of the safety measures adopted by many countries based on the Fukushima Daiichi accident lessons learned was implementing PARs and FCVS in response to severe accidents and mitigation of related consequences. This in turn requires comprehensive analyses on the best performance characteristics and the effectiveness of these mitigation systems under the wide range of accident conditions including not only high but also low probability scenarios with consideration of cliff-edge effects. Therefore, there is an important level of efforts present internationally aimed at improving the models for prediction of phenomena expected to occur in reactor containments during severe accidents including mentioned safety measures. Conventional LP codes are continuously advanced with new models, while the 3D CFD codes are now developed for containment modelling. Both code types require further validations based on new experimental data; usually that requires large scale experiments to consider the impact of physical processes which govern the mixing of containment atmosphere such as, forced convection, natural convection, condensation for an accurate prediction of iodine and aerosol transport and distribution behaviour.

With focus to gain more experimental data and validate safety analysis codes, several new projects have been launched at international level in the aftermath of Fukushima Daiichi accident. Some of these projects (e.g. BSAF, ARC-F) have been aimed to analyse the Fukushima Daiichi accident with an aim to improve severe accident codes and to analyse the accident progression and current status of units 1 to 3 of the Fukushima Daiichi NPP, thus providing useful information for the decommissioning of these units.

At the European level, an in-kind project SAMHYCO-NET in the framework of NUGENIA and an EC funded project named AMHYCO are ongoing at the time of this publication. The projects aim to improve the knowledge concerning on one hand the risk of explosion combustible gases produced during a late-phase severe accident, on the other hand the performance of existing means to prevent this risk and to limit the related consequences. Selected examples of hydrogen related programmes at OECD/NEA and EC levels are as follows:

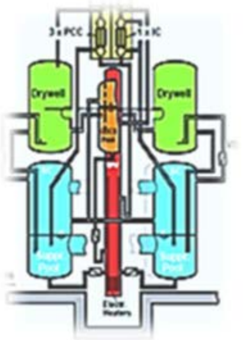
- 2012–2015: Benchmark study of the accident at the Fukushima Daiichi Nuclear Power Station (BSAF)
- 2015–2018: Benchmark study of the accident at the Fukushima Daiichi Nuclear Power Station (BSAF-2)
- 2019~: Examinations inside reactor buildings & containment vessels & water sampling (ARC-F)
- 2014–2018: Severe accident facilities for European safety targets (SAFEST)
- 2016–2019: Thermal-hydraulics, hydrogen, aerosols, iodine (THAI-3)
- 2017–2021: Hydrogen mitigation experiments for reactor safety (HYMERES-2)
- 2012–2018: Access to large infrastructures for severe accidents (ALISA)
- 2017–2020: NUGENIA/SAMHYCO-NET

Experimental test facilities**:** play an important role in understanding different processes or phenomena under severe accident conditions in NPPs. Furthermore, experimental database and a wide spectrum of accident scenarios are required for validation and further development of severe accident analysis tools based on different modelling approaches, such as LP or CFD codes.

Worldwide several large scale experimental facilities, which have now been decommissioned, have played a vital role in hydrogen safety research, e.g. BMC, HDR, NUPEC. Regarding containment safety research in Europe, several existing test facilities like PANDA (PSI, Switzerland), MISTRA (CEA, France), and THAI (Becker Technologies, Germany) have been employed in various OECD/NEA and EC projects covering a wide range of experiments, e.g. separate effect tests (SET), coupled effect tests (CET), and integral tests (IT). Experimental data has been used for International Standard Problems, e.g. ISP-47 (containment thermal-hydraulics) and ISP-49 (hydrogen combustion).

Post-Fukushima Daiichi accident, several new test facilities have either started operation or are in the design phase in Asia Pacific (e.g. Republic of Korea, Japan, China, India). Cooperation between experimental test facilities in Europe and China was realized in the frame of EC-ALISA project. Table 10 provides condensed information about PANDA, MISTRA, and THAI containment test facilities. A good supplement to this publication is the IAEA–TECDOC- 1939 on Post Fukushima Daiichi Accident Developments in the Analysis and Management of Combustible Gases in Severe Accidents in Water Cooled Reactors that provides a list of experimental test facilities employed for hydrogen safety research.

TABLE 10. EUROPEAN TEST FACILITIES FOR NPP CONTAINMENT SAFETY RESEARCH

Facility	**PANDA (PSI)**  	**MISTRA (CEA)** 	**THAI (BT)** 
Reactor type Test specifics	BWR Integral & SET	PWR SET, CET	Generic SET, CET
Phenomena	Thermal hydraulics	Thermal hydraulics	Thermal hydraulics **Severe accidents**
Total volume	460 m^3	100 m^3	60 / 80 m^3
Fluid / medium	He, air, water, steam	He, air, steam	He, air, water, steam **H_2, aerosols, ^{123}I**

6. REVIEW QUESTIONS AND EXERCISES

This Chapter provides review questions (RQ) and working examples (WE).

RQ.1.

Knowing that hydrogen combustion produces heat of 120 kJ/g complete the combustion chemical formula:

+ □ + 120 kJ/g(H_2)

Answer: Section 2.3

RQ.2.

Hydrogen explosion took place during which of these accidents:

a) TMI-2 accident
b) Chernobyl accident
c) Fukushima Daiichi accident

Answer: Section 3.2.2 provides explanation on why hydrogen explosion took place at TMI-2; the rest of this publication speaks of the hydrogen explosion at Fukushima Daiichi NPP.

RQ.3.

Hydrogen gas has a molecular weight of:

a) 2.016 g/mol which is lighter than steam
b) 2.016 g/mol which is heavier than steam
c) Air
d) CO
e) CO_2

Answer: Section 2.1.

RQ.4.

In-vessel sources of hydrogen generation during severe accidents are:

a) Zirconium–steam reaction
b) Steel–steam reaction
c) Boron carbide–steam reaction
d) Uranium–steam reaction
e) Water radiolysis

Answer: Section 3.1.1 explains why the correct answer includes a), b), c) and d).

RQ.5.

Ex-vessel sources of hydrogen generation during severe accidents are:

- **a)** Molten corium–concrete interaction
- **b)** Water radiolysis
- **c)** Corrosion of metallic materials
- **d)** Chemical reaction between corium debris and containment atmosphere
- **e)** Uranium–steam reaction

Answer: Section 3.1.2 explains why the correct answer includes all.

RQ.6.

Explain Shapiro's diagram.

Answer: Review Section 3.2.2.

Shapiro's diagram is used for practical applications. The three sides of the diagram indicate the relative volumetric concentration of air, hydrogen and steam respectively, which are the dominant within the NPP containment atmosphere during severe accidents. The outer curve presents the burn limit, within which a sustainable combustion could occur. A detonation can occur within the inner curve that is the detonation limit. According to Shapiro's diagram, the gas mixture is an inert condition where the combustion cannot occur if the concentration of steam is greater than 55%.

RQ.7.

Explain how the Passive Autocatalytic Recombiner (PAR) works and:

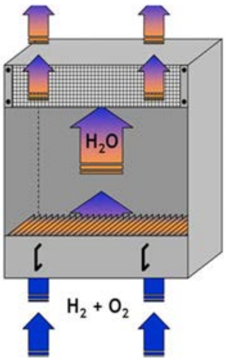
- **a)** Draw the schematics.
- **b)** Write the chemical reactions.
- **c)** Explain the differences for the following three (3) PAR designs.
- **d)** Discuss their radiation resistance.

Answer: Review Section 2.2.

The main recombination processes are:

- Mass transfer of H_2 and O_2 from the main stream through a boundary layer on the outer catalyst surface.
- Transport of H_2 and O_2 to the active centres.
- Adsorption and reaction at the active sites.
- Desorption of H_2O.
- Diffusion of H_2O out of the interior of catalyst to the surface.
- Mass transfer of H_2O from the phase boundary layers into the main stream.

a) PAR schematics:



b) Chemical reactions: Section 2.2.

c) Differences among the provided three PARs:
The main difference between the designs is that both AREVA and AECL designs use catalytic sheet. The NIS design uses catalytic pellet cartridge.

d) Radiation resistance of the PARs from **c)**:
The qualification process for the three PAR designs include the radiation resistance. Their efficiency is not affected by radiation.

RQ.8.

Explain the effects of CO on PAR performance.

Answer:

PARs convert CO into CO_2 following the reaction: $2CO + O_2 = 2CO_2$ or $CO + (1/2)O_2 = CO_2$

CO may behave as poison in some situations, such as lean oxygen mixtures or when the catalytic sheet or pellet cartridge are “cold”.

It is commonly admitted that the threshold temperature is around 100°C. Additional investigations in the framework of the SAMHYCO-NET project are aimed at confirming or revising this value.

RQ.9.

Explain the conditions in the case of molten core–concrete interaction (MCCI) and the parameters that affect the MCCI.

Answer: After the reactor vessel rupture, molten corium will fall into the containment basement. The interaction with the concrete will produce H_2, CO, H_2O and CO_2. Depending on the concrete composition (siliceous or limestone), the quantity of the produced gas is different. More CO production in case of a limestone concrete.

RQ.10.

Explain the conditions, processes, and chemistry behind hydrogen generation in the reactor vessel and containment.

Answer: In-vessel hydrogen generation is divided into early phase and late phase. In the early phase, core uncovering starts and metallic materials such as cladding and absorber, with relatively low melting points, melt and are relocated. The geometry of fuel remains almost intact. In the late phase, the UO_2 fuel melts and dissolves with other metallic materials, forming a ceramic molten mix. As the molten core relocates to a relatively cold area, a crust forms at its surface and causes channel blockage. The crust is continually eroded and reformed as the molten mix relocates to the lower vessel plenum, while particulate debris may be generated during this time.

When the integrity of a reactor vessel fails and molten corium drops into the dry cavity, which consists of concrete, the molten corium–concrete interaction (MCCI) occurs. MCCI is a complicated phenomenon and more so when the molten core is stratified into an oxide layer and a metallic layer. When corium with very high temperature contacts with the cavity surface, exothermic chemical reactions occur between corium materials and gases generated from concrete deposition such as H_2O and CO_2 and concrete becomes ablated.

Find the Sections which describe these aspects?

RQ.11.

Explain the positive and negative effects of spray operation in an atmosphere containing air–steam–hydrogen mixture.

Answer: The spray system activation permits the steam condensation and the mixing of the containment atmosphere leading to the containment pressure reduction, to the increase of the turbulence level and the hydrogen molar fraction in the containment atmosphere. This turbulence level and the hydrogen molar fraction increase may promote flame acceleration in case of combustion.

RQ.12.

Explain positive and negative effects of Passive Autocatalytic Recombiners operation in Light Water Reactor containment.

***Answer*:** PARs are designed to avoid hydrogen accumulation and then the occurrence of dynamic loads in case of ignition. At high concentration of hydrogen, PARs may behave as igniters and initiate a combustion.

WE.1.

To reduce hydrogen concentration inside the containment (CNTM) that has a volume of 10000 m^3, 10 recombiners are used. Suppose that there is 1,000 kg of hydrogen in the CNTM at pressure of 1.5 bar, and temperature of 100 °C. The recombiner rate (*RR*) for each PAR is equal to $A \times CH$, where A = 10 kg/h and *CH* represents hydrogen molar fraction. The CNTM atmosphere contains only air and hydrogen. The hydrogen molar mass is 2 g/mol. The ideal gas constant is R = 8,341 J /mol K.

a) Calculate the initial hydrogen molar fraction (*CH*) inside the CNTM.
b) Calculate the time needed to have a non-flammable mixture.

Answer:

a) Initially, the containment mixture is composed of air–hydrogen which corresponds to a total number of moles as follows:
$N_{total} = (P \times V)/(R \times T)$ = (1.5 bar × 10000 m^3) / ((8,341 J /mol K) × 373K) =
= (1.5 × 10^5 Pa × 10000 m^3) / ((8,341 J /mol K) × 373K) = 4,821,301.67 moles
The initial hydrogen molar fraction is defined as a ratio of hydrogen number of moles N_{H2} per N_{total} as follows:
$CH = N_{H2} / N_{total}$
where N_{H2} = (1000 kg) / (2 g/mol) = 500,000 moles
CH = (500,000 moles) / (4,821,301.67 moles) = 10.37 vol%

b) Non-flammable mixture is reached when hydrogen molar fraction is below CH_{limit} = 4 vol% corresponding to a mass of hydrogen of $M_{H2Limit}$ = 385.7 kg obtained as follows:
$M_{H2Limit} = CH_{limit} \times N_{total} \times$ 2g/mol
The recombined hydrogen mass is then $M_{H2Recom} = M_{H2initial} - M_{H2limit}$ = 1000kg – 385.7 kg = 614.3 kg
The time needed to recombine $M_{H2Recom}$ is then with the given $RR = A \times CH$ = 10 (kg/h) × 10.37 vol% = 1.037 kg/h per each PAR:
$t = M_{H2Recom} / (10 \times RR)$ = 614.3 kg / (10 × 1.037 kg/h) = 59. 2 h.
Therefore, 59.2 h are needed to reach hydrogen molar fraction below 4 vol%.

WE.2.

The NPP engineer has to install PARs in the containment. He knows that there are three available locations:

a) Near ceiling;
b) In annulus (nearby a safety device);
c) In dome region (heavy crane).

But he is not sure which location is preferred one and why?

What will you tell the engineer?

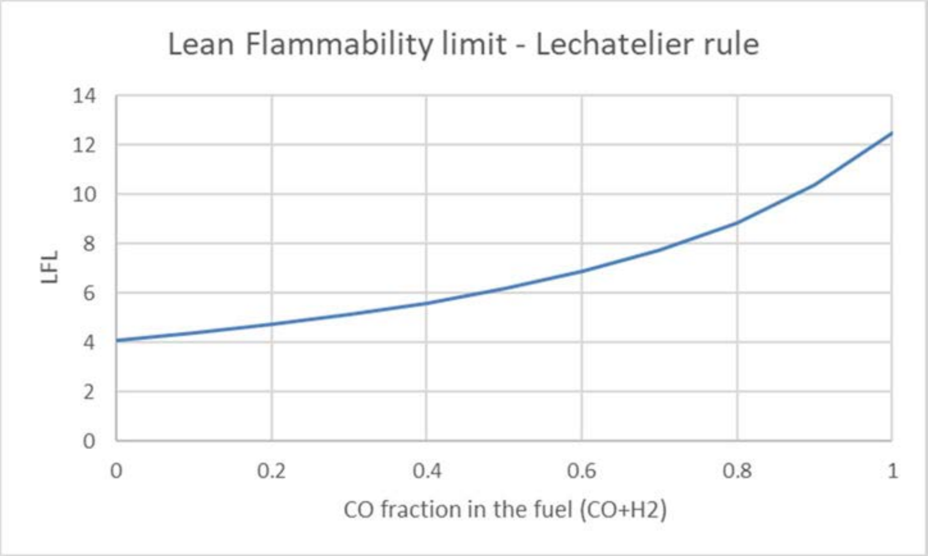
Answer:

The preferred location is the dome region (**c**). The reason is that hydrogen is light gas and move upward when released inside the containment.

WE.3.

In the containment, the atmosphere is composed of 5 vol.% H_2, 12 vol.% CO, and 83 vol.% air. Show if the mixture is flammable or not. The lower flammability of H_2 is 4 vol.% and 12 vol.% for CO.

Answer: Using the Lechatelier rule, the lower flammability limit of CO-H_2-air mixtures could be represented in the following curve:



As the fuel fraction in the mixture is 17 vol% and CO fraction in the fuel is 0.7 than the mixture is flammable.

WE.4.

Hydrogen–steam mixture is injected in the containment (CNTM):

- **a)** in the bottom part of the CNTM;
- **b)** in the dome part of the CNTM.

Show which of the two scenarios will establish stratification and explain why.

Answer: The scenario **b)** with injection in the dome part will easily allow the hydrogen stratification. The hydrogen is lighter than the steam, it will stay in the upper part and then create a stratification.

WE.5.

PAR is operating at full design capacity with catalyst surface temperature on the order of 1000°C. What is the source term issue if CsI aerosols pass at this moment through PAR?

Answer: In contact of the hot catalytic surface, CSI molecule can be thermally decomposed and release gaseous iodine.

WE.6.

Explain the importance of R&D focus on stratified atmospheres in view of H_2 risk.

Answer: In case of stratification, local accumulation of hydrogen can pose a risk of combustion. Most of the R&D programs, performed in the past, dealt with gas distribution in homogenous conditions. As the most critical conditions leading to high pressure loads in case of combustion is related to stratified atmosphere configurations, the recent R&D programs are focused on hydrogen combustion behaviour in stratified conditions. Thus, complex phenomena governing hydrogen stratification build-up and break up were investigated in the OECD THAI and SETH projects. The flame propagation in stratified conditions was investigated in OECD THAI and still an issue and is investigated in the ongoing SAMHYCO-NET and AMHYCO programs.

WE.7

Explain the difference between active and passive hydrogen recombiners. Which one is expected to perform better in SBO (station blackout) scenario?

Answer: Active recombiners operation needs power. In case of SBO accident, the loss of power will unable these systems to operate. Whereas, PARs require no power for operation and exothermic reaction on the catalytic surface heats the plates and activates natural convection driven circulation of the gases.

7. CONCLUDING REMARKS

In the course of a severe accident in a water cooled reactors, hydrogen is produced in amounts of up to several hundred kilograms and released into the containment. Its deflagrative combustion can pose a threat to safety components and in a worst scenario to the containment integrity. Maintaining NPP containment integrity is of fundamental importance to avoid fission product release to the environment as a comparison of the severe accidents in TMI-2, Chernobyl and Fukushima NPPs make obvious.

Even through the present publication has been intended to provide basic knowledge on hydrogen safety aspects, the information is sufficient to offer an overall picture of hydrogen behavior and the related risks during severe accident conditions. A wide range of topics have been covered including hydrogen generation, propagation, combustion and mitigation techniques of relevance to water cooled power reactors. The fundamental information is complemented with practical examples using main outcomes or references of ongoing international activities relevant for hydrogen behavior in NPPs.

One specific use of information provided in this publication is to offer a basic framework for workshops, education, or expert training courses related to hydrogen behavior under severe accident conditions. As per needs of Member States, contents can be further elaborated without missing the most important elements required to train and educate on the risk informed decision making on hydrogen safety related aspects.

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GLOSSARY

This glossary compiles relevant reference terminology from the IAEA Safety Glossary (2018 Revision)).

accident conditions. Deviations from normal operation that are less frequent and more severe than anticipated operational occurrences.

ⓘAccident conditions comprise design basis accidents and design extension conditions.
ⓘExamples of such deviations include a major fuel failure or a loss of coolant accident (LOCA).

containment. Methods or physical structures designed to prevent or control the release and the dispersion of radioactive substances.

ⓘAlthough related to confinement, containment is usually used to refer to methods or structures that perform a confinement function in facilities and activities, namely preventing or controlling the release of radioactive substances and their dispersion in the environment.

design basis accident. A postulated accident leading to accident conditions for which a facility is designed in accordance with established design criteria and conservative methodology, and for which releases of radioactive material are kept within acceptable limits.

early release of radioactive material. A release of radioactive material for which off-site protective actions are necessary but are unlikely to be fully effective in due time.

emergency response. The performance of actions to mitigate the consequences of an emergency for human life, health, property and the environment.

fission product. A radionuclide produced by nuclear fission.

fuel element. A rod of nuclear fuel, its cladding and any associated components necessary to form a structural entity.

ⓘCommonly referred to as a fuel rod in light water reactors.

model. An analytical or physical representation or quantification of a real system and the ways in which phenomena occur within that system, used to predict or assess the behaviour of the real system under specified (often hypothetical) conditions.

model validation. The process of determining whether a model is an adequate representation of the real system being modelled, by comparing the predictions of the model with observations of the real system.

monitoring. The measurement of dose, dose rate or activity for reasons relating to the assessment or control of exposure to radiation or exposure due to radioactive substances, and the interpretation of the results.

nuclear accident. Any accident involving facilities or activities from which a release of radioactive material occurs or is likely to occur and which has resulted or may result in an international significant transboundary release that could be of radiological safety significance for another State.

protection and safety. The protection of people against exposure to ionizing radiation or exposure due to radioactive material and the safety of sources, including the means for achieving this, and the means for preventing accidents and for mitigating the consequences of accidents if they do occur.

risk assessment. Assessment of the radiation risks and other risks associated with normal operation and possible accidents involving facilities and activities.

safety analysis. Evaluation of the potential hazards associated with the operation of a facility or the conduct of an activity.

safety function. A specific purpose that must be accomplished for safety for a facility or activity to prevent or to mitigate radiological consequences of normal operation, anticipated operational occurrences and accident conditions. (See SSG-30 [53]).

safety limits. Limits on operational parameters within which an authorized facility has been shown to be safe.

safety system. A system important to safety, provided to ensure the safe shutdown of the reactor or the residual heat removal from the reactor core, or to limit the consequences of anticipated operational occurrences and design basis accidents.

safety system settings. Settings for levels at which safety systems are automatically actuated in the event of anticipated operational occurrences or design basis accidents, to prevent safety limits from being exceeded.

severe accident. Accident more severe than a design basis accident and involving significant core degradation.

transport. The deliberate physical movement of radioactive material (other than that forming part of the means of propulsion) from one place to another.

uncertainty analysis. An analysis to estimate the uncertainties and error bounds of the quantities involved in, and the results from, the solution of a problem.

validation. The process of determining whether a product or service is adequate to perform its intended function satisfactorily.

ABBREVIATIONS

AICC	Adiabatic Isochoric Complete Combustion
BR	Blockage Ratio
BWR	Boiling water reactor
CFD	Computational fluid dynamics
DBA	Design basis accident
DDT	Deflagration to detonation transition
EC	European Commission
FA	Flame Acceleration
FCVS	Filtered containment venting system
FL	Flammability limit
IAEA	International Atomic Energy Agency
IT	
LFL	Lower flammability limit
LP	Lumper parameter
LOCA	Loss of coolant accident
LWR	Light Water Reactor
MCCI	Molten core concrete interaction
NPP	Nuclear power plant
PAR	Passive autocatalytic recombiner
PHWR	Pressurized heavy water reactor
PWR	Pressurized water reactor
TMI	Three mile island
SET	Separate Effect Tests
UFL	Upper flammability limit
WCR	Water cooled reactor
WOG	Westinghouse owner group
WWER	Water water energetic reactor

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Meetings

National Training Workshop on Modelling Physics and Technology of Hydrogen Management in Severe Accidents, 15−19 April 2019, Beijing, China

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