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Hydrogen Program at AECL

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ABSTRACT

Hydrogen combustion in containment could challenge containment integrity and pose a threat to critical accident management equipment. Demonstration of the effectiveness of hydrogen mitigation features (igniters or recombiners) is required to provide assurance that accident consequences are bounded.

AECL has a well-established program for performing hydrogen dispersion analysis for CANDU design basis accidents, using the GOTHIC code, which has been extensively validated against data from AECL large-scale experimental facilities, and the DDTIndex code for assessing the potential threats to containment integrity resulting from Deflagration-Detonation Transition (DDT) or from flame acceleration in non-uniform post-accident H_2/air/steam mixtures.

Wet-proof Passive Autocatalytic Recombiners (PARs) were developed and are now marketed by AECL for mitigation of hydrogen inventory in containment, especially for the long-term period in loss-of-coolant accident scenarios, for use in CANDU and PWR reactor designs.

The Large-Scale Vented Combustion (LSVC) test facility was used to systematically quantify effects of key thermodynamic and geometric parameters affecting combustion pressure development in vented rooms under conditions relevant to deliberate ignition, and for environmental qualification testing of PARs. The facility has good control of initial thermodynamic conditions, is sufficiently large to capture the effects of scale and is geometrically similar to rooms in containment buildings (i.e., flat walls and square corners). The experiments are performed under prototypical thermodynamic conditions relevant to CANDU containments, and produce data suitable for code validation.

This paper gives a brief overview of AECL's hydrogen R&D program, with focus on recent multi-chamber PAR/atmosphere interaction experiments performed in the LSVC facility. The program philosophy has been to establish sound theoretical and experimental foundations for the development of prediction methods for post-accident CANDU containment response as well as to provide sufficient code validation data.

1 HYDROGEN RISK IN CANDU AND PWR REACTORS

Following a postulated Loss-Of-Coolant Accident (LOCA) in a CANDU or PWR reactor, hydrogen (or deuterium in the case of heavy-water reactors) could be produced in the core and released into containment through the break in the Primary Heat Transport System (PHTS) or Reactor Coolant System (RCS) along with high-temperature steam. The production of hydrogen in the core results in the short term from Zircaloy fuel elements that are inadequately cooled, reacting exothermally with the steam present in the RCS, and in the long term from water radiolysis and metal corrosion. In containment, the hydrogen/steam mixture will mix with air forming a combustible mixture. Mechanical and thermal loads from hydrogen combustion could threaten containment integrity and thus lead to unacceptable fission product release to the environment.

Thermodynamically, the reaction of fuel sheath (cladding) with steam, followed by hydrogen combustion in containment, is equivalent to oxidation of the fuel sheath by containment air. In this process hydrogen provides the storage and transport mechanism for some of the energy to move from the core into containment. The amount of hydrogen produced and thus the energy transported depends on the fuel temperature and the availability of steam to sustain the Zr-steam reaction. The risk to containment is a function of this energy and its rate of release. Hydrogen risk mitigation means to limit this energy release rate in containment. Overpressure in containment can be prevented if hydrogen remains unchanged or is consumed slowly, for example using PARs, in small burns ignited near the mixture flammability limit, or by establishing a standing diffusion flame through autoignition of the hot steam/hydrogen mixture right at the break. In the latter case, while overpressurization of containment in not a concern, the heat load from the standing flame on safety equipment will become the dominating safety concern..

While in most PWR regulatory jurisdictions the failure of the Emergency Core Cooling System (ECCS) or Emergency Coolant Injection (ECI) system is not considered within the reactor design basis and therefore falls within the severe accident regime, the CANDU reactor design basis has traditionally included LOCA/LOECI accidents. As the vendor of CANDU reactors, AECL has developed a good understanding of hydrogen phenomena through a comprehensive R&D program on hydrogen generation, transport, combustion and mitigation in nuclear reactors over the past 30 years. Irrespective of the present or past treatment of the hydrogen risk within or outside the design-basis accident (DBA) regime, the direction of most reactor regulators to risk-based methods requires current nuclear reactor vendors and utilities to address the hydrogen risk quantitatively over the entire range of possible containment conditions.

A comprehensive discussion of the hydrogen risk, relevant phenomena, and available experimental data and computer codes relevant to nuclear reactors can be found in two recent OECD State-of-the-Art Reports (Ref. [1,2]).

2 RISK MITIGATION STRATEGIES

Risks to (1) containment integrity and (2) post-accident equipment survivability can be addressed in a number of ways. Both types of risks result from the potential combustion (or recombination) of hydrogen with containment air, and the resultant pressure and temperature increase. Ideally, pressure and temperature excursions can be precluded, if the hydrogen is not allowed to burn. A second acceptable mitigation strategy is to remove the hydrogen, either through recombination or controlled combustion(s), which limit the pressure and temperature increase within acceptable limits, i.e. within the containment envelope design pressure and equipment qualification limits.

Mitigation strategies, such as pre-inerting and active containment venting, are not discussed here, because they have not been part of CANDU hydrogen risk mitigation strategies

2.1 Dilution

Not all mixtures of hydrogen and air will burn and even those that do burn under some conditions may not under different conditions. Several ingredients are necessary to produce sustained hydrogen combustion: sufficient concentrations of fuel (hydrogen or deuterium) and oxygen, a transport mechanism to bring the reactants together (sufficient pressure, turbulence, molecular diffusion), and ignition source of sufficient energy (spark, hot surface or fluid, flame, shock wave). Since the latter two ingredients, gas transport and an ignition source, cannot be precluded in a post-accident containment atmosphere, the only controllable ingredients are the hydrogen and oxygen concentrations.

Some BWR reactor designs control oxygen concentration by providing an inert (nitrogen) containment atmosphere, thus addressing the hydrogen combustion risk. Short of fully inerting the containment atmosphere, the oxygen concentration cannot be controlled in PWR and CANDU containment designs.

Under pressures relevant to containment, a hydrogen/air mixture will not burn if the hydrogen concentration is below 4%. Therefore, dilution is an acceptable hydrogen risk mitigation strategy, if it can be shown that the hydrogen concentration remains below 4% under accidents for which containment integrity must be maintained. In the past, single-unit CANDU stations have relied on this strategy for DBA's (including LOCA/LOECI), based on the limited hydrogen source term from the in-core Zr-steam reaction combined with a large containment volume available for dilution. However, an increased hydrogen source term can be expected under certain postulated severe accidents, and dilution by itself can no longer be relied on as the sole hydrogen mitigation strategy.

2.2 Deliberate Ignition

Theoretically, a flammable hydrogen-air-steam mixture can remain unburned for an indefinite time, in the absence of an ignition source. However, in containment there are a number of potential ignition sources: sparks from electrical equipment or static buildup, flames from conventional fires, hot surfaces (glow plug igniters, PAR plates, fuel fragments), focusing of shock waves from energetic local events, e.g. from a steam explosion, and even autoignition of the hot hydrogen/steam jet escaping from the RCS break. Therefore, it is plausible that a burn would eventually occur if a flammable mixture is present and it would be non-conservative to assume that a flammable mixture will not burn because no known ignition source is present. On the other hand, it would also be non-conservative to assume that the mixture would burn as soon as it becomes flammable and before it grew to a size and concentration where its deflagration could damage containment.

In contrast to the above accidental (or random) ignition sources, spark or glowplug igniters provide an intentional source of ignition. These devices are in use in several CANDU reactors and have been investigate in detail to ensure reliable performance under a variety of conditions. AECL has studied the performance of different types of igniters and the effect of hot surfaces in general as a source of ignition (Ref.[3]). Results showed that steam could raise the ignition temperature (about 75-100ºC at 30%), and had a stronger effect than other diluents, initial gas temperature had little effect and pressure had a significant effect. Figure 1 shows the effects of pressure and steam concentration for both hydrogen and deuterium.

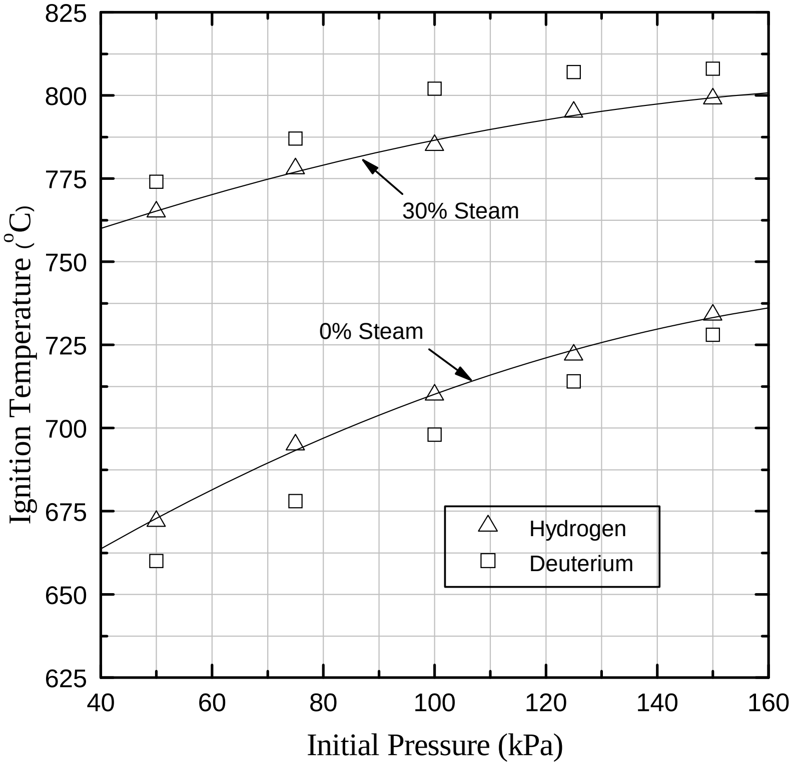

Figure 1: Effect of Steam and Pressure on Minimum Surface Ignition Temperatures for H_2 and D_2 Mixtures (from Ref. [3])

The progress of the combustion also depends on the location of the ignition source. For example, experiments in AECL's Large-Scale Vented Combustion (LSVC) facility in a single-chamber geometry with venting to atmosphere have indicated that peak pressure significantly depends on the igniter location (near vent, central, or far from vent, Ref [4]). Depending on the hydrogen concentration, either far-vent or central igniter locations resulted in the highest peak pressures. The effect of igniter location in a vertical direction is also important, because of the different upward and downward flame propagation characteristics. As a result of buoyancy, a flame will propagate upward at a lower concentration than downward; for hydrogen in air these limits are 4% and 9%, respectively. Consequently, an igniter near the bottom of a well-mixed hydrogen cloud of between 4 and 9% hydrogen) would be more effective in removing the hydrogen from containment than one located near the top. On the other hand, if the cloud tends to be stratified, enriched with hydrogen near the top, an igniter near the top is preferred. In any case, when igniting hydrogen/air mixtures near the flammability limit, it is possible and quite likely, that not all the hydrogen is removed. Experiments done in multi-compartment geometries at AECL have shown that below 8% hydrogen in air, incomplete combustion can be expected, with varying degrees of unburned hydrogen depending on igniter location. Therefore, the location of igniters in nuclear containments becomes important for lean mixtures.

Deliberate ignition is a hydrogen risk mitigation strategy that attempts to prevent unacceptable local or global hydrogen concentrations that, if allowed to ignite, would result in unacceptable pressure and temperatures. It does this by forcing hydrogen clouds to burn near their flammability limit, and thus can prevent accumulation of hydrogen in containment. It relies on proven post-accident igniter reliability, judicious igniter location, following the above-described principles, combined with validated hydrogen mixing calculations.

2.3 Recombination

Passive Autocatalytic Recombiners (PARs) enable the oxidation of hydrogen to proceed without combustion, at hydrogen concentrations below the flammability limit and at temperatures below the ignition temperature. The heat of the exothermic reaction generated in the PAR creates a natural convective flow through the recombiner, eliminating the need for pumps or other active devices to bring hydrogen to the catalyst surface. The hot hydrogen-depleted steam-air-hydrogen mixture exits from the top of the recombiner and pulls in hydrogen-rich cool mixture into the bottom inlet, which sustains the reaction at the plates and the convective flow through the PAR as long as the ambient mixture contains sufficient hydrogen to sustain the reaction (typically 0.5 to 1.0%). This convective loop promotes strong mixing in the room where the PAR is installed.

PARs are designed to operate in the range of 1% to 7% hydrogen in air. The thin catalyst plates and relatively wide spacing dissipate heat and enable operation well into the flammable range of mixture compositions. Above 7% hydrogen the plates become hot enough to possibly cause a hot surface ignition of the hydrogen air mixture. In as-new condition the AECL recombiner self-starts typically between 1% and 2% hydrogen in a steam-saturated atmosphere at room temperature (Ref. [5]). The low-temperature self-start threshold may temporarily increase as a result of exposure to contaminants, but once started, the PAR operates at the design hydrogen removal capacity and recovers its as-new self-start limit.

Similar to deliberate ignition, PARs remove containment hydrogen at low concentrations, well below levels where containment integrity is threatened. It also requires judicial placement of the recombiners in locations where hydrogen is expected.

3 HYDROGEN MITIGATION R&D AT AECL

A prerequisite to the successful implementation of all hydrogen mitigation strategies discussed above - dilution, deliberate ignition, and recombination - is a good understanding of the mixing behaviour of the hydrogen that is released into containment.

3.1 Hydrogen Dispersion

When active measures that promote gas mixing in containment are present, such as fan coolers or containment sprays, the assumption that the released hydrogen mixes quickly and completely throughout most of the available volume in containment, can be easily supported. However, in the absence of such forced mixing, the dispersion of hydrogen from the break in the break room and eventually throughout the containment relies on weaker forces, such as buoyancy-induced pressure gradients resulting from concentration and temperature (gas mixture density) differences.

The simulation of gas mixing in an enclosure or between rooms driven by buoyancy differences requires multi-dimensional modeling (1D, 2D or 3D depending on the geometry complexity). Validation of this phenomenon requires detailed gas concentration, and heat transfer measurements, which are scale dependent. Therefore, the gas mixing tests for GOTHIC code validation were performed at as large a volume as practical with scenario relevant parameters in order to reduce scaling uncertainties.

AECL has a well-established program for performing hydrogen dispersion analysis for CANDU design basis accidents using the GOTHIC code, which has been extensively validated against data from AECL large-scale experimental facilities. Two such facilities are the Large-Scale Gas Mixing Facility (LSGMF, located at Whiteshell Laboratories) and the Large-Scale Containment Facility (LSCF, located at Chalk River Laboratories, shown in Figure 2).

Results from a large number of tests (using helium as a simulant for hydrogen) in both facilities showed that air entrainment into the helium plume results in rapid mixing along the plume axis. This was particularly noticeable for bottom injection tests, where dilution of the injected helium prevented any strong stratification of helium inside the facility. For example, air entrainment between the injection location (near the floor) and the ~1000-m^3 facility ceiling (about 10 m above the floor) into the pure-helium plume resulted in dilution to ~2-3% helium, and even after 10 minutes of injection the helium concentration did not exceed 5% anywhere in the room. When helium was slowly injected closer to the room ceiling, the helium “pooled” and high concentrations were measured near the ceiling. A pre-stratified steam layer had a significant effect upon gas mixing, reducing the gas-mixing rate until the local water vapour concentration gradients decreased. GOTHIC multi-dimensional models predicted these effects quantitatively with acceptable accuracy.

Currently, multi-compartment mixing experiments are being conducted to extend the validation base of GOTHIC to more complex geometries and scenarios.

Figure 2: Large-Scale Containment Facility (LSCF) used to perform helium/air/steam mixing experiments

3.2 Hydrogen Combustion

AECL has completed a comprehensive experimental program investigating the hydrogen combustion behaviour from near-flammability limits up to stoichiometry at various scales. Numerous publications are available in the literature (Refs. [3-7]). The focus has been on these relatively low concentrations in support of the deliberate-ignition strategy employed in some CANDU stations. To illustrate the complexity of the hydrogen combustion in the lean regime, Figure 3 (taken from Ref. [4]) shows the peak pressure variation with hydrogen concentration for various igniter locations in the LSVC facility. It is interesting to note that whereas the peak pressure increases monotonically with hydrogen concentration for near-vent and central ignition, peak pressure rise with far-vent ignition exhibits non-monotonic behaviour.

An abridged summary of the investigations covered by AECL’s hydrogen combustion research program is as follows:

1. Flammability limits and deflagration behaviour of hydrogen-oxygen-diluent gas mixtures for a wide range of hydrogen content (near-flammability limits up to stoichiometric) as a function of composition, initial thermodynamic parameters, turbulence, ignition mode, confinement scale (ranging from bench- to large-scale) for conditions relevant to post-LOCA, including vented combustion in multi-volume configurations.
2. Intense radiation fields and hydrogen isotope effects on hydrogen ignition/flammability properties.
3. Hydrogen combustion-acoustic coupling phenomenon studies.
4. Flame acceleration, DDT and detonation properties of hydrogen gas mixtures.
5. Diffusion flame properties for hydrogen-air-steam compositions.

A limited subset of AECL's hydrogen combustion experiments was used to validate GOTHIC for calculating slow, vented combustion events and moderate obstacle-induced flame acceleration. Figure 4 shows a result from simulating a series of vented combustion experiments in the LSVC facility with ~10% hydrogen and 0-30% steam. GOTHIC slightly underpredicts the mitigating effect of steam but generally calculates a reasonable overpressure (peak pressure above initial pressure) and impulse (pressure-time integral).

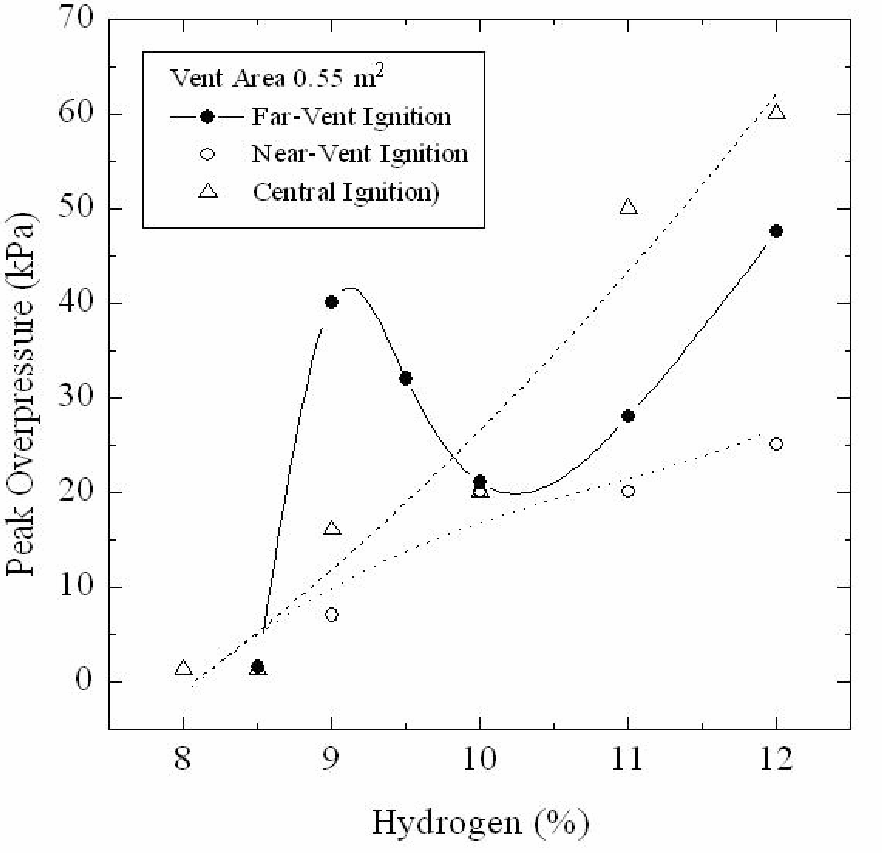

Figure 3: Effect of Steam and Pressure on Ignition Temperature (from Ref. [3])

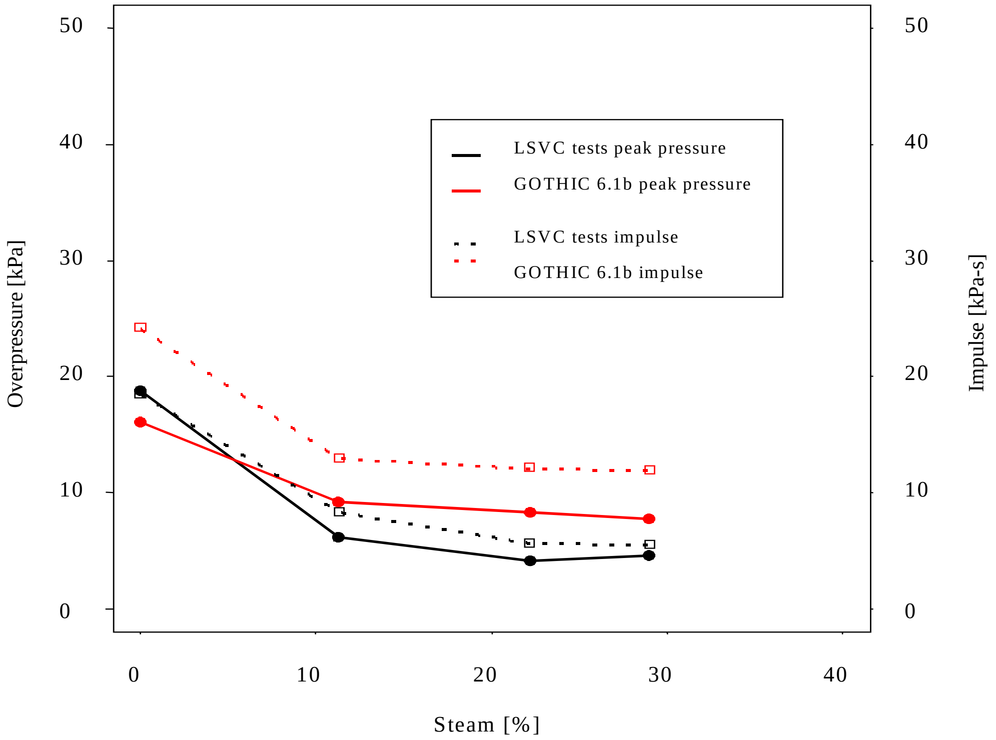

Figure 4: Comparison of Overpressure Between Experimental Results and GOTHIC Simulations

3.3 AECL Recombiner

The AECL wet proof autocatalytic recombiner is a passive hydrogen removal device that uses a platinum catalyst to convert hydrogen and oxygen into water vapour and heat. Designed for use in post-LOCA conditions where hydrogen is present in containment, it was designed, developed, constructed and qualified by AECL [5]. The requirements for recombiner design and the extensive qualification test program were specified to bound foreseeable conditions in CANDU and PWR large dry containments. The qualification test program was designed to demonstrate that the PARS can perform their intended function during and after being subjected to the following conditions: Thermal and radiation aging under normal conditions for the lifetime of the station, exposure to a design basis earthquake, and exposure to expected normal and post-accident contaminants present in containment, including volatile organics. Performance testing also included tests at high pressure, low oxygen, with spray (clean and contaminated water) and with fuel aerosols. The original AECL proprietary catalyst formulation (AECL Type 89-24) has the most complete scope of qualification testing and the alternative formulations (AECL Type 99-11 and Type 99-17) have improved resistance to some volatile organic compounds and molecular iodine and otherwise similar performance.

The AECL recombiner, shown schematically in Figure 5, consists of an open-ended rectangular stainless-steel 304/304L box with an attached cover and gratings. It has a nominal inlet flow area of 0.2 m^2. The top cover and gratings provide physical protection to the internal elements from sprays or missiles. Inside the box, 31 flat rectangular catalyst elements are arranged parallel to the direction of gas flow. These elements are spaced approximately 2 cm apart to promote optimal convective flow, and can be easily accessed by removing or pivoting the top cover.

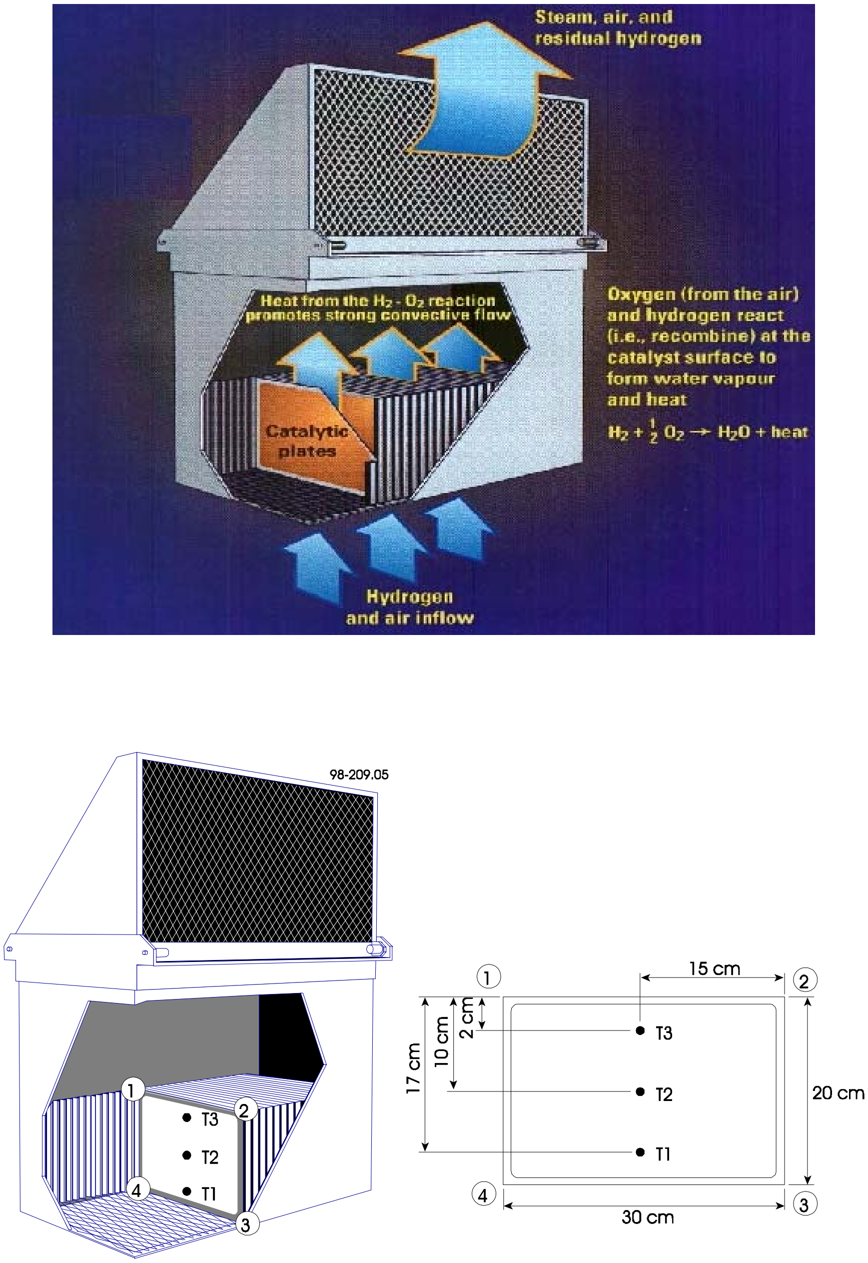

Figure 5: Schematic of AECL Wet-Proof Recombiner and Details of Thermocouples on Plate 16, as used in the Multi-Chamber PAR Mixing Tests

PARs create a convective flow within the compartment they are located, and have been shown to cause hydrogen mixing and thus effective depletion within a single compartment. A short test program was recently completed to investigate how the action of a single PAR affects the mixing and depletion in a configuration of multiple, interconnected chambers (two and three chambers). Openings existed between chambers and hydrogen concentration was monitored in all chambers. The test results are used to demonstrate that a code, such as GOTHIC, can adequately simulate this mixing between volumes with and without a PAR. In a typical reactor building installation, not all rooms and not all regions of a larger room will be equipped with PARs.

A schematic of the LSVCTF, which has been used for PAR qualification testing, is shown in Figure 6. The total volume of the test facility is ~120 m^3 (L/W/H = 10/4/3 m) Nominally, the front chamber is 60 m^3, and each of the rear chambers is 30 m^3. The horizontal partition separating the top and bottom rear chambers can be removed to create two chambers, each of 60m^3, and the centre (vertical) partition can be removed to make a single chamber of 120 m^3.

Figure 6: Large-scale Vented Combustion Test Facility configured for Multi-Chamber PAR Tests (rear top/bottom chambers on left, front chamber on right)

Variations in openings between the different chambers, PAR box/hood orientation, and facility temperature were examined. For the three chamber tests, vents could be opened between the top and bottom rear chambers, between the top rear chamber and the front chamber, between the bottom rear chamber and the front chamber, or a combination thereof. Two or three vents were opened for each of the tests. For the two chamber tests, an upper or lower horizontal vent between the front and a rear chamber was used. Two tests were conducted in just the front chamber (baseline), and two counterpart tests for the two- and three-chamber configurations were carried out to show reproducibility.

Depending on the chamber configuration (number of chambers, vent configuration), gases in the interconnected chambers may not thoroughly mix, creating a non-uniform final distribution in the different chambers. Once the recombiner self-stops, the hydrogen remains in the other chambers, giving an indication of the degree of mixing during the test. In most tests, the hydrogen in all chambers was adequately removed by the PAR, except for several tests in which one of the rear compartments was a “dead-end” volume, connected to the front chamber containing the PAR through a vent below the PAR elevation. Gas mixing was impeded by this configuration. A dead-end volume with a vent above the PAR location (i.e. the rear top chamber) was adequately mixed by the action of the recombiner.

4 REMAINING KNOWLEDGE GAPS AND INTERNATIONAL COLLABORATIONS

The hydrogen program at AECL has spanned the last 3 decades and is coming to a successful completion, in as much as experimental investigations into hydrogen dispersion, deflagration, detonation and recombination, and related design-basis code qualification is concerned. A few gaps in the understanding of important phenomena remain, and these are briefly outlined below:

1. Validation of hydrogen burn models in integral containment codes (such as GOTHIC) for lean mixtures where incomplete combustion is expected, and where the differentiation between upward and downward propagation is important. AECL is currently extending the validation using existing in-house data and also participating in the OECD THAI project.
2. Modeling of PARs in complex scenarios, interaction of PARs with containment flow fields and multi-compartment geometries. The recently completed PAR tests at AECL are an extension of existing databases. Participation in SARNET WG12-2 PAR benchmarks addresses some fundamental scenarios of interest in PAR operation and will hopefully conclude in a relevant validation exercise, possibly using AECL data.
3. Quantifying the impact of local hydrogen burns on equipment qualification (EQ).
4. Qualifying new AECL PAR designs, supporting nuclear station specific implementation and installations to support the non-nuclear hydrogen economy.

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