QUALIFICATION OF PASSIVE AUTOCATALYTIC RECOMBINERS FOR POST-LOCA HYDROGEN MITIGATION IN CANDU STATIONS

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1. INTRODUCTION

One possible consequence of a loss of coolant accident in a CANDU® reactor is that hydrogen may be released into the containment at the start of the accident. The worst case scenario is the loss of coolant accident coincident with a loss of emergency core cooling (LOCA/LOECC) for which flammable mixtures of hydrogen and air may occur a few hours following the accident. In the longer term, radiolysis and corrosion can also increase the hydrogen concentration. Presently, natural and forced mixing are relied upon to dilute the hydrogen to non-flammable concentrations in the large air volume of the containment. In some designs, ignitors are installed to burn hydrogen where local concentrations exceed flammable limits. Passive autocatalytic recombiners have been designed for long-term hydrogen control and to improve margins for short term hydrogen management. They operate by combining hydrogen with oxygen at a catalyst surface to form water. The heat of reaction at the catalyst surface changes the water to steam, and creates a natural convective flow through the recombiner. This flow acts as a pump to bring more hydrogen through the recombiner and enhances mixing inside the containment. This paper describes the qualification program for the AECL hydrogen recombiner and presents the results from selected performance tests.

2. THE AECL RECOMBINER

The AECL recombiner is designed for compactness and ease of engineering into containment. The design consists of an open-ended rectangular box (32 cm × 62 cm × 52 cm) with an attached cover and gratings (see Figure 1). The cross-sectional area of the open ends is 0.2 m^2. Inside the box, 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. This box with the catalyst elements is the basic recombiner module. The mounting frame is made from rectangular steel tubing to provide secure support for the recombiner module and an interface to containment structures. The optional cover and gratings provide physical protection to the internal elements from sprays or missiles.

The catalyst used in the recombiners described in this work is a proprietary AECL formulation developed specifically for application in nuclear containments [1,2]. The catalyst has a high catalytic activity for hydrogen oxidation, is not deactivated by water vapour or steam, and is specially formulated for operation over a very wide range of temperatures. It is wet-proofed using a proprietary formulation, in which the platinum catalyst is dispersed within a porous support structure of synthetic silica zeolite, offering a high surface area. The zeolitic structure of micropores acts as a selective molecular sieve. Water molecules cannot enter the matrix, but hydrogen and oxygen are able to diffuse to the active surface of the platinum for the recombination reaction to occur. The catalyst operates at temperatures up to 1000 K without loss of wet-proofing or catalytic activity and is unaffected by high radiation exposures. It has shown resistance to poisoning by anticipated containment gases [3].

The operation of the AECL hydrogen recombiner is completely passive: it requires no support systems (electrical power, instrument air, cooling water), no controls, and is self-starting in the presence of hydrogen and oxygen. Self start, as evidenced by catalyst heating and initiation of convective flow, has been demonstrated in early tests at 1% hydrogen in air, room temperature, and 100% relative humidity [3]. This low temperature start-up in condensing atmospheres is viewed as the most challenging condition for wet-proofing effectiveness. Cold start-up is a vital performance requirement in CANDU containments, which contain engineered air-cooling systems and where long-term hydrogen control is required after containment atmospheres have cooled. Tests comparing recombiner performance at different locations within a 3-m×4-m×10-m test chamber show that recombiner location does not have a strong effect on capacity [3].

The recombiners would be distributed throughout the reactor building, from the lower level floor (above the water flood level), to the upper areas of the building. Recombiners are protected from water sprays by gratings and an angled (45 degree) cover. The recombiners would be anchored to the internal structure of the reactor building using supports made of rectangular carbon steel tubing. When installed in containment, the recombiners would be part of the hydrogen control system, classified as a Group 2 safety-related system in CANDU 6 stations and as a Group 1 subsystem in the CANDU 9 design. Their safety function would be to reduce and maintain the hydrogen concentration in containment below the flammability limits in the long term (beyond the first 24 hours) after design basis accidents.

3. QUALIFICATION REQUIREMENTS

The "AECL 1996" standard recombiner is being qualified for use in existing CANDU 6 stations (Gentilly-2, Point Lepreau, Wolsong 1 and 2/3/4) and provisions are being made for the extended station lifetime of 40 years for Qinshan 1 and 2 and for future CANDU 9 stations. The hydrogen recombiners are not required to operate during normal service conditions. However, the recombiners must be able to operate for the mission time of 12 months after being subject to: 1) the normal service conditions inside the reactor building, for the lifetime of the reactor; 2) the abnormal service conditions after a design basis accident; and 3) a site design earthquake 24 hours or later after the accident.

Recombiners would be located in an environment where periodic testing of the catalyst plates is possible. The catalyst plates are the only components of the recombiner subject to aging, as the rest of the recombiner consists of a stainless steel open-ended box. Temperature, periodic testing, and radiation are the only anticipated aging mechanisms. These aging mechanisms are not expected to significantly affect catalyst performance. However, they are included in the qualification tests.

During the lifetime of the station and after a site design accident, the recombiners would be exposed to temperatures ranging from 0° - 140°C, atmospheres containing up to 8% hydrogen and 100% relative humidity, and pressures ranging from just below atmospheric to 2 atmospheres. These conditions bound all of the stations considered for qualification. The recombiners must also remain functional after a Site Design Earthquake. They must also maintain their structural integrity during and after a Design Basis Earthquake, to avoid becoming a potential source of damage to other DBE qualified components in containment.

In order to choose the number of recombiners required in containment and to analyze their effectiveness in reducing the hydrogen levels, the recombiner performance must be defined. The performance of the recombiners is defined using the following characteristics:

1. The self-start limit, or the hydrogen concentration above which the recombiner will start recombining hydrogen;
2. The self-stop limit, or the hydrogen concentration below which the recombiner stops recombining hydrogen. (This limit is different from the self-start limit because once the recombiner is hot from the catalytic reaction, it can recombine lower concentrations of hydrogen when it is starting cold.);
3. The capacity, or the recombination rate, in kilograms hydrogen per hour.
4. The single pass efficiency, which is the percentage of hydrogen which is recombined into water in one pass through the recombiner.

Performance tests are conducted under a number of reference conditions which are representative of the most challenging conditions for the recombiner. The most challenging condition for the recombiner is cold (~10°C) and wet (100% relative humidity), with a low hydrogen concentration (1%). Further performance tests are conducted for a broader range of conditions and to study the sensitivity of the recombiner performance to a number of parameters. These tests proof test the design and address potential questions from regulators and future customers. They examine the effects of the following parameters:

- mixture composition below and above the lower flammability limit as well as beyond the steam inerting threshold.

- reduced and increased ambient temperature
- hydrogen combustion
- steady hydrogen injection over an extended period of time
- a reduced number of catalyst plates in the recombiner
- recombiner location within the test chamber
- potential fouling or poison agents (for example, water spray, aerosols, fog, steam, dust, cable fire soot, iodine, etc.)

4. QUALIFICATION TEST PROGRAM

4.1 Test Sequence

Functional testing is performed both before and after accelerated thermal aging and radiation, to quantify the effects of aging on recombiner performance. The test sequence is as follows:

1. Initial inspection and documentation
2. Baseline functional tests
3. Radiation exposure
4. Thermal aging
5. Cycle aging
6. Post-aging functional and performance tests

Seismic testing is performed on an unaged recombiner because aging will not affect the recombiner's ability to withstand seismic testing. For the seismic tests, the sequence is as follows:

1. Baseline functional tests
2. Seismic tests
3. Post-seismic functional tests

4.2 Test Facility and Instrumentation

Environmental qualification testing of the AECL recombiner is performed in the Large Scale Vented Combustion Test Facility (LSVCTF) at Whiteshell Laboratories [4]. The LSVCTF is a 120 m^3 (10 m long, 4 m wide, and 3 m high) structural-steel enclosure temperature-controlled for operation at atmospheric pressure and ambient temperatures up to 140°C. The facility has systems for the controlled addition of hydrogen, steam, and inert gases. The composition of hydrogen, oxygen, and steam inside the test chamber is measured using an industrial process mass spectrometer with a high speed rotary sampling valve.

Temperatures and gas composition are monitored at the inlet and outlet to the recombiner and at locations at mid-height, ceiling level, and floor level inside the test chamber. Three thermocouples are attached to a catalyst plate, evenly spaced in the direction of gas flow. The recombiner is located at the centre of the chamber for most of the tests, and near the end wall for the location-effect tests. The recombiner module is mounted inside the test chamber by suspending it from the ceiling with cables.

4.3 Test Procedure

The test procedure for environmental tests performed in the Large Scale Vented Combustion Test Facility is described briefly as follows:

- The test chamber is completely sealed.
- The test chamber is heated to the required test temperature.
- Hydraulic mixing fans are turned on to mix incoming gases with the air already in the chamber. (Mixing is almost instantaneous.)
- Steam is added to the chamber until 100% humidity (or the desired steam concentration) is achieved.
- Hydrogen is added to the chamber until the target mixture is achieved and verified by the gas analyser output. This mixture composition begins to change almost immediately because the recombiner begins operating as soon as hydrogen reaches it.
- Gas addition is stopped and the mixing fans are turned off so that the natural convection loop driven by the recombiner is allowed to control the flow movement.
- The mass spectrometer measures and records the change in gas composition in the chamber and the data acquisition system records the thermocouple output.
- When the gas composition stops changing (usually at 0.5% H_2), the mixing fans are turned on to confirm mixture homogeneity. The final gas composition is recorded, thermocouple sampling is halted and the test is ended.

5. ENVIRONMENTAL FUNCTIONAL AND PERFORMANCE TESTING: SELECTED RESULTS

5.1 Self Start

The cold, wet self-start behaviour at 1% H_2 is shown in Figure 2. At time = 0.0 min., the hydrogen addition begins. Shortly afterwards, the catalyst elements were exposed to the hydrogen-steam-air mixture, and recombination began, as evidenced by the drop in hydrogen concentration at the recombiner outlet. The mixing of gases in the test chamber was good, as indicated by the agreement of bulk gas and inlet samples. Th recombiner converted approximately 70% of the hydrogen in the test chamber in 100 minutes. The concentration dropped from 1% to 0.3% before the recombiner stopped operating.

5.2 Self-Stop

A typical self-stop concentration is 0.3% hydrogen (see Figure 2). Tests at different initial conditions have consistent self-stop values. Once the catalyst is heated by the catalytic reaction, it can recombine lower concentrations than when the catalyst is cold. Since the heat of reaction produced at a specific hydrogen concentration is always the same, the recombination rates at any concentration (all other factors being the same) are independent of the initial hydrogen concentration of the test.

5.3 Efficiency

The single pass efficiency of the commercial model recombiner is approximately 55%, as determined from inlet and outlet hydrogen concentrations. It is noted that the recombiner was designed to optimize overall capacity, not efficiency. Higher single pass efficiencies are possible, but at the expense of flow velocity and overall capacity. A typical analysis is shown in Figure 3, for a 4% H_2 test.

5.4 Capacity

The capacity of the recombiner is calculated from the change in hydrogen concentration of the bulk gas in the test chamber. The capacity, 1-kg/hour/m^2 inlet area/%H_2, changes linearly with hydrogen concentration, and is independent of the initial concentration. Previous tests have shown that the capacity scales linearly with the inlet area of the recombiner [3]. The capacity of the recombiner, calculated from several tests, is shown in Figure 4.

6. CONCLUSIONS

The intended function for passive autocatalytic recombiners is to reduce and maintain the hydrogen concentration in containment below the flammability limits in the long term after loss of coolant accidents. The recombiners must be able to operate for the mission time of 12 months after being subject to: 1) the normal service conditions inside the reactor building, for the lifetime of the reactor; 2) the abnormal service conditions after a design basis accident; and 3) a site design earthquake 24 hours or later after the accident.

Qualification tests are conducted under a number of reference conditions which are representative of the most challenging conditions for the recombiner. The test facility for environmental performance and qualification testing is the Large Scale Vented

Combustion Test Facility at Whiteshell. Performance tests confirm recombiner self-start behaviour in cold, wet conditions and demonstrate recombiner capacity in typical post-LOCA environmental conditions. Once started, the removal capacity normalised to the inlet cross-section area of the recombiner scales approximately linearly with the hydrogen concentration.

7. REFERENCES

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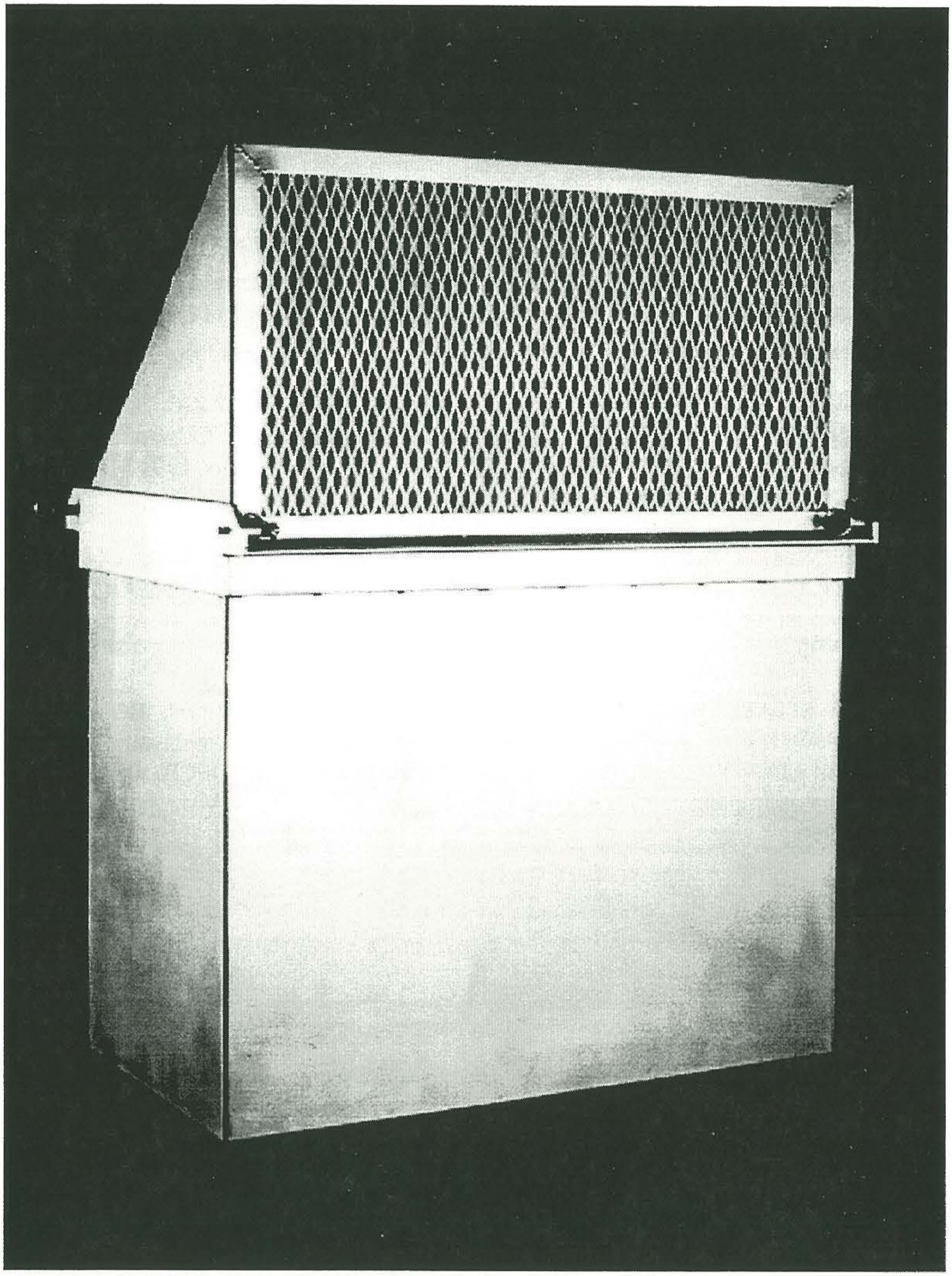

Figure 1.   AECL Hydrogen Recombiner

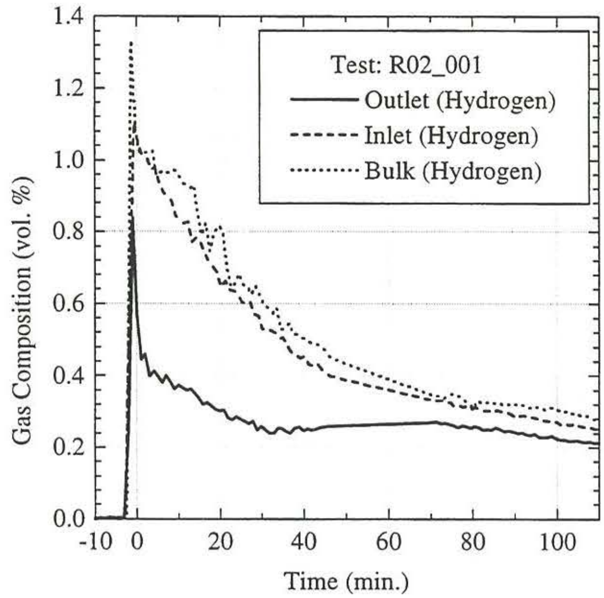

Figure 2. Cold, Wet Self-Start at 1% Hydrogen, 25 C.

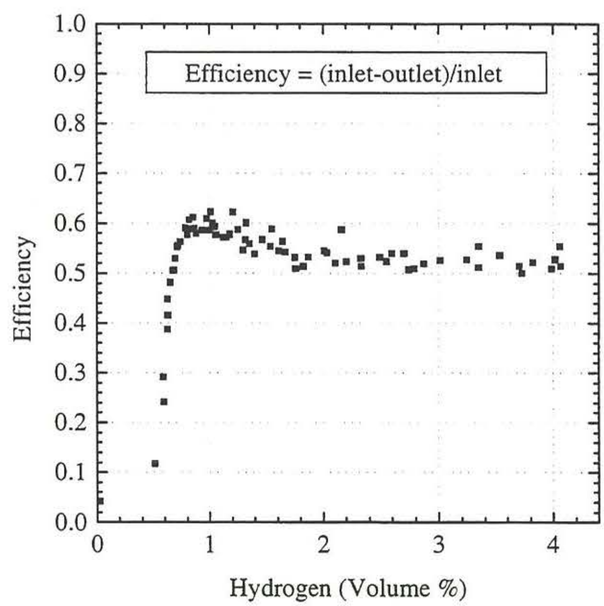

Figure 3. Single-Pass Efficiency of Hydrogen Recombiner.

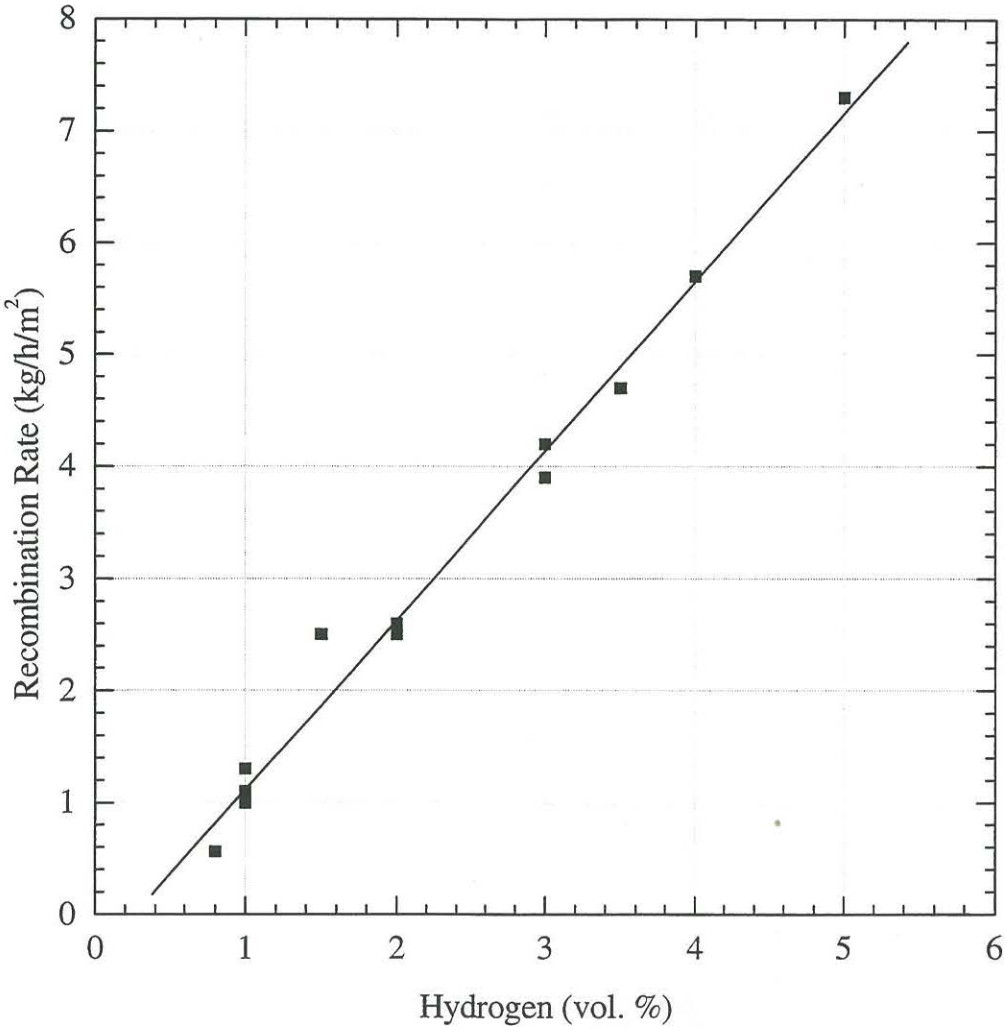

Figure 4. Capacity of Hydrogen Recombiner.