THE 4TH INTERNATIONAL TOPICAL MEETING ON
NUCLEAR THERMAL HYDRAULICS, OPERATIONS AND SAFETY
April 6-8, 1994, Taipei, Taiwan

PASSIVE AUTOCATALYTIC RECOMBINERS FOR COMBUSTIBLE GAS CONTROL IN ADVANCED LIGHT WATER REACTORS

U. Wolff and G. Sliter
Nuclear Power Division
Electric Power Research Institute
P.O. Box 10412
Palo Alto, California 94303
(415) 855-7945

ABSTRACT

A key aspect of the worldwide effort to develop advanced nuclear power plants is designing to address severe accident phenomena, including the generation of hydrogen during core melt progression (metal-water and core-concrete reactions). This design work not only resolves safety concerns with hydrogen, but also supports the development of a technical basis for simplification of off-site emergency planning.

The dominant challenge to any emergency planning approach is a large, early containment failure due to pressure excursions. Among the potential contributors to large and rapid increases in containment pressure is hydrogen combustion. The more improbable a containment-threatening combustion becomes, the more appropriate the argument for significant emergency planning simplification. As discussed in this paper, catalytic recombiners provide a means to passively and reliably limit hydrogen combustion to a continuous oxidation process with virtually no potential for containment failure in passive advanced light water reactors (ALWRs).

INTRODUCTION

Passive ALWRs are under development by General Electric, Westinghouse, U.S. and international utilities, the Electric Power Research Institute (EPRI), and the Department of Energy.[1] Basic design principles include (in addition to safety) design simplicity, maintainability, and preference for passive components that do not have moving parts and do not rely on active power sources or support systems. These principles have led to adoption of the passive autocatalytic recombiner (PAR) as the preferred approach required by U.S.-utility and international ALWR participants for controlling combustible gases in ALWRs.[2]

PARs perform their function passively, sucking atmospheric gases containing hydrogen and oxygen at the bottom of a steel box-like device and blowing out water vapor from recombination at the top. Inside the device, recombination occurs at the surface of porous palladium-coated ceramic pellets used as a catalyst. PARs are self-starting and self-feeding, even under cold and wet conditions. Buoyancy of the hot gases they create sets up strong convective flow currents that promote mixing of combustible gases in a containment. Hydrogen in PWRs (or oxygen in inerted BWRs) begins to be recombined as soon as it is introduced. The recombination rate of a number of PARs distributed throughout a containment volume keeps the average concentration of hydrogen (or oxygen) below regulatory limits based on representative release rates. Since PARs have no moving parts and require no external energization, they require no operational procedures and are easily maintained. This is projected to lead to greater life-cycle cost-effectiveness, which is also a goal of passive ALWRs.

This paper gives a brief description of the design and qualification of PARs and how they would be implemented in *passive* ALWRs. (If this application appears sufficiently cost-effective, PARs could be considered for *nonpassive* ALWRs or operating plants.) The paper then contrasts the PAR approach with conventional hydrogen control systems based on igniters.

Although the PAR concept is applicable to control of combustible gases released during both design basis accidents and severe accidents in both PWRs and BWRs, this paper concentrates on the PAR system design for mitigation of severe accidents in PWRs. (A PAR system designed to meet combustible gas control requirements for severe accidents will assure that combustible gas concentrations for design basis accidents will remain well below flammability limits at all times. The Appendix gives a brief description of PAR application in a BWR.)

DESIGN AND QUALIFICATION OF PARs

The candidate design of a PAR system used for the ALWR feasibility study discussed in this paper is the pelletized design developed and qualified in Germany by the NIS Company in cooperation with Degussa (catalyst supplier), the Battelle Institute (test laboratory), and the Technical University, Munich (analysis). Its development has been sponsored by the German utility RWE Energie, which is a participant in the ALWR Program.[3] (Note that there are other catalytic recombiner designs that could perform the functions; these other designs are also candidates for selection by ALWR plant designers.)

The NIS /RWE PAR device shown in Figure 1 is a "molecular diffusion filter" (not the more conventional fixed-bed particle filter, in which gases are forced through the interstitial spaces between catalyst particles). The device consists of 88 flat, rectangular cartridges containing a total of about 30 kg of spherical catalyst pellets having 1-cm-wide open flow channels between the cartridges. The immense surface area of the palladium-coated outer layer of the porous ceramic pellets acts upon diffused gas molecules, while heavier particles or aerosols in the atmosphere flow through the open channels with little plugging of the pellet surfaces. The gas flow is sucked in at the bottom of the device, recombined while passing through the flow channels, and funneled into a chimney blowing the heated gas through a square hole at the top.

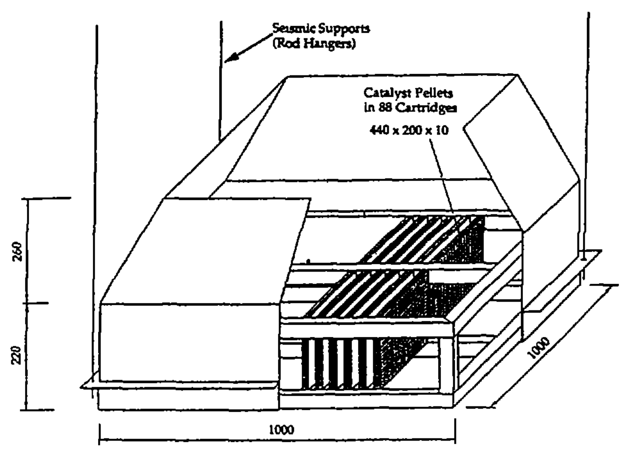

Figure 1 - Drawing of the Prototype PAR Device (Dimensions are in cm.)

Design optimization, extensive determination of performance characteristics, and qualification of the PAR device has been realized through a three-phase test program performed by NIS and Battelle[4] that included screening tests of various catalysts, model tests to characterize performance, and full-size prototype tests in a multicompartment model containment.

A best fit to the model and prototype test data gave the empirical curves of PAR depletion rate as a function of hydrogen concentration of the gas entering the device in Figure 2. These values of depletion rate were used for the benchmarking mixing and distribution code calculations and the simplified depletion analysis of a containment discussed in the next section. Note that on the basis of test results designed to examine the effect of poisons, such as iodine and carbon monoxide, on the performance of the PAR catalyst system it is justified to conclude that they do not significantly reduce the performance of PARs given by the curves in Figure 2.[2,3]

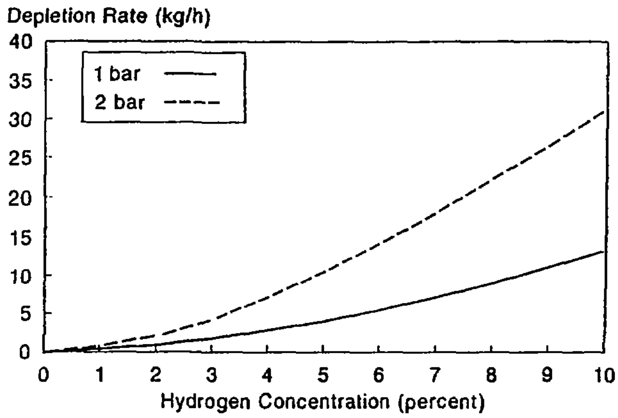

Figure 2 - Experimentally Determined PAR Depletion Rate as a Function of Hydrogen Concentration
(1 bar = no steam, 2 bar = 50% steam)

APPLICATION OF PARS IN ALWR CONTAINMENTS

A simplified, conservative analysis has been used to investigate the feasibility of applying the PAR concept to plants. The analysis assumes a uniform gas distribution averaged throughout the containment volume and, therefore, treats only the global aspect of hydrogen control (i.e., ensuring that global concentrations do not exceed regulatory limits, thereby ensuring that the containment can perform its function even during severe accidents). More sophisticated thermal-hydraulic computer codes (such as the COBRA-NC/GOTHIC code) will eventually be used by designers to perform detailed analyses of specific plant applications. These analyses will treat both global and local aspects of hydrogen control. Regarding local aspects, the overall approach must prevent any situation producing a local detonation (or flame acceleration near detonation) that compromises the safety functions of structures or equipment in a local region or compartment. In the following paragraphs, we (1) describe the simplified analysis and results of a PAR plant application, (2) discuss the factors that justify use of the simplified analysis for this feasibility study of global performance, and (3) discuss the factors and measures that the PAR concept relies upon to address the possibility of local detonations.

Conservative Simplified Analysis

The first step in the analysis is to specify the total amount and rate of hydrogen release into the containment. The Code of Federal Regulations[5,6] gives clear guidance concerning generated/released quantities of hydrogen to be considered. In accordance with 10CFR50.34(f), which addresses the concern of global detonation, the total hydrogen quantity generated/ released during a severe accident must be calculated from a reaction of 100% of the active fuel clad material. Also, the average uniform concentration in the containment building shall never exceed 10% hydrogen.

An envelope of the hydrogen release rates as determined in PRA best-estimate severe accident analysis for a variety of scenarios was estimated to perform the simplified design analysis.[2] It was assumed that all hydrogen is released within one hour and twenty-five minutes (50% of total hydrogen quantity in the first 10 minutes, 25% in the next 35 minutes, and the rest in the next 50 minutes).

To illustrate the approach used for the simplified analysis, a sketch of the assumed hydrogen release and calculated hydrogen depletion for a severe accident scenario in a typical ALWR PWR containment is shown in Figure 3. The upper curve shows the hydrogen concentration in the AP600 containment that would result from release in accordance with the release assumption described above without any hydrogen depletion. The conservative assumption of no steam content is made. The lower curves give the calculated hydrogen concentration for different numbers of PAR devices installed in the containment. For the first 10 minutes, the simplified analysis assumes there is no depletion. This is a conservative treatment of the startup time during which hydrogen may have not reached some PARs and other PARs may be heating up, not yet working at full efficiency. Recombination is assumed to begin only after 10 minutes. From this time on, the depletion is calculated using a time-step-average hydrogen depletion rate (from Figure 2 assuming no steam content and ambient pressure) determined iteratively. The number of PAR devices determines the peak hydrogen concentration. This peak concentration (which is allowed to be as high as 10% in a best-estimate analysis) occurs between about one half and one and a half hours. With one PAR per 3000 m^3 of containment volume, hydrogen recombination prevents the volume average concentration from exceeding 10%. Following termination of hydrogen release, the step calculation is continued. Several hours after the start of the accident, the atmosphere is inerted by the action of the PARs. Within 24 hours, only small amounts of hydrogen would remain.

The analysis shows that with just enough PARs deployed to keep global concentrations below 10 vol%, the atmosphere could be flammable at most only for a few hours. During this relatively short period, inadvertent ignition is acceptable because important systems, structures, and components are designed for the loads resulting

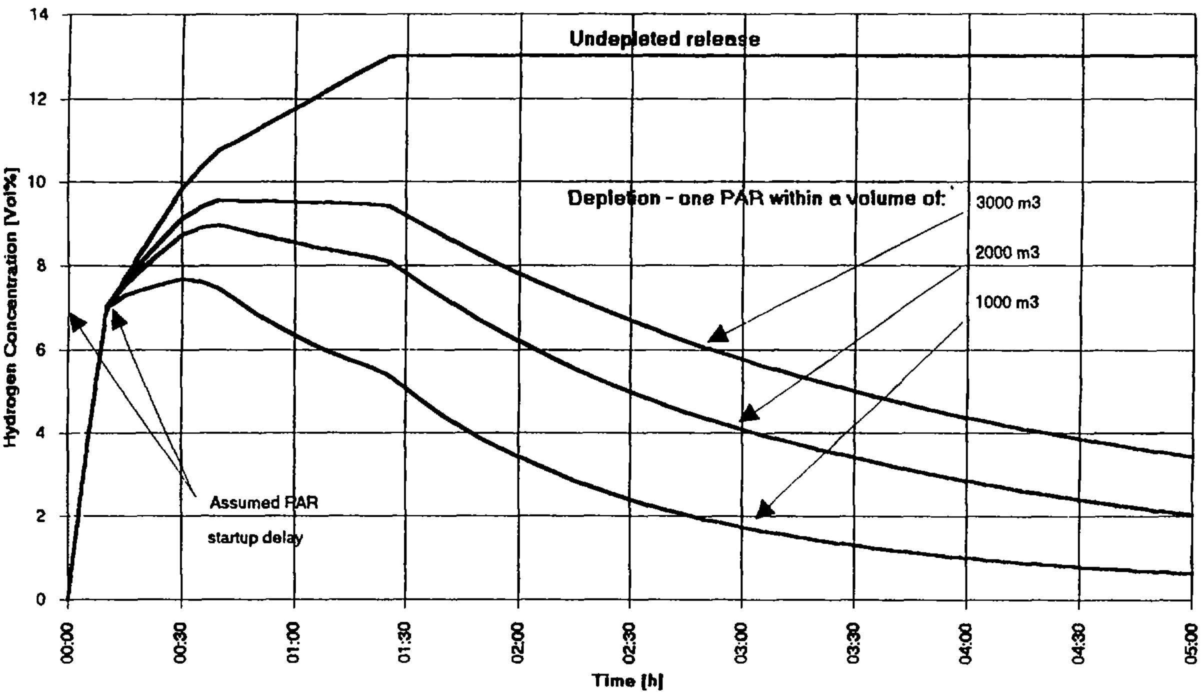

Figure 3 - Sketch of a Typical Assumed Hydrogen Release and Calculated Hydrogen Depletion for a Severe Accident Scenario in a PWR Containment.

from a hydrogen burn of such a gas mixture. However, it is important to recognize that, in view of the several sources of conservatism in this simplified analysis, it is likely that hydrogen concentration will remain below even the flammability limit at all times for many severe accident scenarios. The following reasons are the basis for this expectation:

- It is conservative to assume that the total hydrogen would be generated in less than two hours (50% in only 10 minutes). Any stretching of the generation over longer times would reduce maximum concentrations. This statement is valid even for a less probable scenario in which fuel would exit the vessel and have a chance to react with concrete leading to additional generation of hydrogen. Such generation would occur later when most of the original release is already recombined.

- It is conservative to assume zero steam content, because (1) a large fraction of the water inventory will be released in the form of steam prior to any hydrogen generation; for each 10% of steam the maximum hydrogen concentration reduces by ≈1%; a steam content ≥50% inerts the atmosphere for any concentration of combustible gases, and 2) it is virtually impossible (especially with the high release rates assumed) for the molten core to function as a 100% effective hydrogen generator, chemically reacting with all steam before the steam has a chance to leave the zone of the molten core.

The simplified analysis of hydrogen depletion was applied to the containment of the Westinghouse AP600 PWR. For a containment volume of 48,144 m^3 and a total hydrogen release of 635 kg, 20 PAR devices were found to control the peak volume average hydrogen concentration to no greater than 8.5%.[a]

Uniform Global Concentrations

Here, we review features of hydrogen release and mixing in the presence of PARs to justify the assumption of uniform global concentrations inherent in the simplified analysis.

The momentum of released gases together with the prevailing natural circulation produced by large temperature gradients will initiate gas mixing (these global temperaure gradients result from high temperatures associated with the accident and passive cooling of the containment dome in the AP600). Hydrogen will reach the nearest PAR devices within a few seconds following release. This can be recognized by noting that the maximum distance of the nearest device to any potential release location is less than about 10 to 15 m^1 and the release velocity is expected to be well above 10 m/sec. (For AP600, the gas exit velocity would reach ≥60 m/sec for a hydrogen release rate of 0.5 kg/sec, even assuming a fairly large release area of 1 ft^2.)

After starting, the PAR device also acts as a "passive fan" strongly supporting gas mixing in the containment atmosphere. For example, twenty PAR devices exposed to air containing 4% hydrogen create a gas flow of about 18,000 m^3/hr. For 10% hydrogen, twenty PAR devices create a circulation of about 30,000 m^3/hr. This is the same order of magnitude as the capacity of one of the two circulation fans used to ventilate the AP600 during normal operation—about 50,000 m^3/hr (31,000 scfm). Even at 1% hydrogen concentration, the twenty devices create a flow of 6,700 m^3/hr. Thanks to this strong self-mixing of PARs, in addition to the strong thermal currents present in an accident scenario, the distribution of gases in the containment can be considered to stay practically uniform starting shortly after the release of hydrogen and for as long as hydrogen and oxygen continue to be present in the atmosphere. Once established, the strong mixing also minimizes the extent of the local region at the point of reactor coolant system leak or break that by definition will have high concentrations of hydrogen.

The feature of good mixing to produce a reasonably uniform distribution of hydrogen has been demonstrated experimentally in the Battelle multicompartment test facility and theoretically using the validated COBRA-NC/GOTHIC code. Since all analysis is based on conservatively considering noncondensable gases only, steam condensation could not lead to any situation that is not covered by the design and needs no further consideration. Note also that the overall depletion rate of PARs distributed fairly uniformly throughout the containment free volume has little dependence on actual hydrogen distribution because regions of lower concentrations with slower depletion are balanced by regions of higher concentration with faster depletion. This gives us confidence that the simplified analysis is an adequate means for estimating the required number of PARs.

Local Concentrations

Although the simplified analysis described above made the conservative assumption that there is no depletion during a 10-minute startup stage for the PARs, the assumption would be over conservative if applied for *estimating concentrations in an individual compartment*, since a PAR near the break location would see hydrogen immediately and begin depletion. On the other hand, the fact that there are regions of the open volume of the containment and its compartments with higher concentration of hydrogen is inherent in the simplified analysis assumption of a uniform *average* concentration. That is, the simplified analysis does not address potentially high local concentrations of hydrogen. Here, we review factors and

[a] The larger-volume containment (90,200 m^3) of a nonpassive PWR, the ABB-CE System 80+, was found to need 40 PARs to keep the peak concentration to 8.1% with a total hydrogen release of 1,126 kg. Note that neither this result nor the result for the AP600 correlates with the curves in Figure 4 because slightly different parameters were used in the calculations for the figure.

measures used by the PAR approach to resolve the concern for the possibility of local detonations.

Hydrogen release into the free volume of the containment building will occur at the location of the primary system break which originally caused the accident and/or at a location such as a relief valve where the primary system was opened for depressurization. Since such a release could contain pure hydrogen produced by the metal/water reaction, it is not possible during dilution to avoid either a local region with hydrogen concentrations well above 10 vol% or a standing flame in the vicinity of hydrogen entry.

In such locations, a stream of hydrogen and steam will be released and will dilute into the free volume of the containment building driven by the momentum of the release with support from natural and PAR-enhanced convection. For many accident scenarios, the release stream would contain 50% or more of steam, so it would not be flammable. If the steam fraction of the release is less than 50%, flammable gas concentrations may exist in local areas for the short time it takes dilution into the free volume to take place. Effective mixing and minimization of the size of regions of high concentrations at entry locations will be promoted by designing compartments with boundaries having a large percentage of their surface open.

Although the initial release of steam-rich mixtures of hydrogen into air may lead to an inert atmosphere, steam condensation may eventually produce a flammable mixture in a compartment. The likelihood of this is reduced by the reduction of hydrogen both by mixing and diffusion to adjacent free volumes and by PAR recombination during the inert period.

Although, as discussed above, it is highly unlikely that pure hydrogen with no steam is released, let us examine such a case. For small release rates, the arrangements of PAR units within individual compartments will keep the average local hydrogen concentration below 10%. For large release rates, the compartment would be quickly inerted due to oxygen purging by the inrushing hydrogen. Oxygen that would enter later due to diffusion or mixing would be recombined by the PAR unit or units, avoiding conditions that would allow local detonation. It is only relatively unlikely intermediate release rates of uninerted hydrogen that could both locally overload PARs and allow oxygen to be present.

Thus, although detonable local regions (average local hydrogen concentration greater than 10%) cannot be totally excluded, they are extremely unlikely to occur. If analysis shows that they could occur during a significantly great time interval, two ALWR design measures minimize the probability of local detonations. First, designs will avoid compartment configurations (e.g., long, narrow regions with obstacles and small jet openings) that could support flame acceleration sufficiently to form a local detonation. Second, designs will minimize possible sources of inadvertent ignition in the compartment, with special attention to avoid the high-energy ignition sources needed to initiate detonation.

The above factors and measures, supported by the results from detailed thermal-hydraulic analyses of PAR effectiveness, will probably lead to the conclusion that the probability of a local detonation is acceptably low. If there are still local regions where such a conclusion cannot be made with sufficient certainty the following two alternative measures can be explored.

The first alternative would be to design such regions [e.g., the condensation pool (IRWST) gas space in a PWR] to have what we will call "PAR venting." This involves closing the compartment to the open containment volume except for a vent or vents with PAR devices installed in them. In this design approach, combustible gases in the compartment are controlled in the following way. The vents require all gas transfer (out or in) to pass through the PARs. An additional PAR installed within the compartment is sufficient to control the hydrogen concentration for small release rates. For intermediate and large hydrogen release rates, rapid inerting of the compartment would occur mainly from oxygen purging through the vent as pressure buildup in the compartment is relieved. This would be assisted to some extent by recombination in the PAR device within the compartment. After achievement of inerting, hydrogen would be released through the vent and dilute into the open containment volume. Following termination of hydrogen generation, the hydrogen content of the compartment would recombine slowly with oxygen entering through the PAR in the vent.

The second alternative can be applied whether or not PAR venting is used. This alternative relies on the ability of structures and components in and around a region or compartment to withstand a local detonation. Analysis and/or test data would be used to predict the short-duration pressure spike from a local detonation and evaluate its effects to ensure that it would not produce an unacceptable level of structure or equipment damage or failure. The successful outcome of such an evaluation will be enhanced by (1) the limited energy available in a detonation in a localized region, (2) the acceptability of large deformations so long as the function of affected structures in an accident situation is not compromised, and (3) the small number of equipment items that (a) are needed to perform a safety function during a severe accident, (b) are not rugged enough to survive a local detonation, or (c) cannot be relocated to a region with an acceptably low probability of local detonation.

COMPARISON OF COMBUSTIBLE GAS CONTROL SYSTEMS

As a result of lessons from the Three Mile Island accident, operating nuclear power plants with relatively small containments have been backfitted with additional systems to control combustible gases produced during and after design basis and severe accidents. In contrast with the PAR approach, conventional AC-powered igniter systems in existing U.S. power plant applications rely on deliberate local or global deflagration to limit global hydrogen concentration below detonable limits for severe accident scenarios. As a noninerted mixture of hydrogen is released into the containment, igniters burn hydrogen as soon as flammable mixtures reach an igniter location. If the released mixture is inerted with more than a 50-vol% concentration of steam, an igniter can begin to burn hydrogen only after steam condensation produces a noninerted mixture.

Most of these systems employ electrical igniters inside the containment and thermal recombiners inside or outside the containment. Because of their limited capacity, the thermal recombiners are used only to prevent hydrogen produced by radiolysis during a design basis accident from reaching flammable concentrations (about 5 vol%) and to assist in the removal of unburned hydrogen after an accident. Igniters are needed for noninerted containments in the event that the hydrogen produced by metal/water reaction in a degraded core accident would exceed the flammability limit. Controlled burning at or above this limit aims at preventing global concentrations from exceeding detonable limits (a volume average >10 vol%). Such detonations could compromise the integrity of the reactor containment structure.

PARs function even for hydrogen concentrations below the flammability limit or in hydrogen-rich steam-inerted mixtures. This attribute reduces the probability of producing flammable concentrations of hydrogen. However, if flammable concentrations are reached, inadvertent ignition from electrical devices or hot surfaces may lead to local or global deflagration. Therefore, the containment and other important equipment are designed for resulting mechanical and thermal loads from deflagrations of hydrogen concentrations as great as 10 vol%.

PARs limit global hydrogen concentrations below detonable limits during severe accidents (and below flammable limits during design basis accidents), entirely eliminating the need for AC- or DC-powered igniters and AC powered thermal recombiners. PARs remove residual hydrogen after an accident. They are less expensive to purchase and maintain than systems relying on two types of devices.

SUMMARY AND CONCLUSIONS

The PAR approach relies mainly on distributed recombination to prevent global detonation. PARs remove almost all hydrogen after the accident. Local detonation is addressed by (1) demonstrating its acceptably low probability (steam inerting, strong mixing, compartment design, and minimization of ignition sources), (2) redesigning potentially problematic compartments to be isolated from the open containment volume with PAR vents, or (3) by assuring survivability (i.e., that compartment structures and equipment can accommodate a local detonation).

In conclusion, the PAR approach is viewed as meeting regulatory requirements for hydrogen control while being simpler, easier to maintain, and more cost-effective than conventional igniter/thermal recombiner systems. Thus, the PAR approach is more in keeping with the design philosophy of the ALWR program. Upon acceptance of the PAR approach by the U.S. Nuclear Regulatory Commission and proper application (including detailed thermal-hydraulic and, if needed, structural analysis by designers), the issue of hydrogen control during severe accident scenarios in ALWRs will have a cost-effective resolution. This resolution is a key element of the technical basis for simplification of off-site planning.

REFERENCES

1. Advanced Light Water Reactor Utility Requirements Document, Executive Summary, EPRI (1990).

2. "Qualification of Passive Autocatalytic Recombiners for Combustible Gas Control in ALWR Containments," EPRI ALWR Program (1993).

3. U. WOLFF, "Control of Hydrogen Concentration in Reactor Containment Buildings by Using Passive Catalytic Recombiners," 2nd ASME/JSME International Conference on Nuclear Engineering (ICONE), San Francisco, California (1993).

4. U. BEHRENS, M. SEIDLER, and U. WOLFF, "Hydrogen Mitigation using Catalyst Modules," Proceedings of a Workshop on Hydrogen Behavior and Mitigation in Water-Cooled Nuclear Power Reactors, Brussels, March 1991, 286 (1992).

5. 10CFR50.34(f), "Additional TMI-Related Requirements," Subparagraph 50.34.(f)(1)(xii).

6. 10CFR50.44, "Standards for Combustible Gas Control System in Light-Water Power Reactors."

APPENDIX—APPLICATION OF PAR CONCEPT IN INERTED CONTAINMENTS

For preinerted containments as in BWRs the time dependence of hydrogen release during severe accidents as well as the total quantity released are not of interest for the design of combustible gas control systems. Any massive hydrogen release will only further reduce the relative oxygen concentration well below flammability limits. The only task for PAR devices is to consume the slowly generated radiolytic oxygen (or oxygen from other sources, if any) to assure that flammability conditions will never be reached.

Application of the conservative simplified analysis for the passive ALWR BWR (General Electric SBWR) showed that 2 standard size plus 8 quarter size PAR devices would assure that following the release of hydrogen, the oxygen concentration will be well below 4 vol%. The PARs can also be employed to remove residual post-accident hydrogen by the controlled gradual injection of oxygen into the containment.