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# CONTROLLING HYDROGEN BEHAVIOR IN LIGHT WATER REACTORS

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## ABSTRACT

In the aftermath of the incident at Three Mile Island Unit 2 (TMI-2), a new and different treatment of the Light Water Reactor (LWR) risks is needed for public safety because of the specific events involving hydrogen generation, transport, and behavior following the core damage. Hydrogen behavior in closed environments such as the TMI-2 containment building is a complex phenomenon that is not fully understood. Hence, we present an engineering approach for prevention of loss of life, equipment, and environment in case of a large hydrogen generation in an LWR. A six-level defense strategy is described that minimizes the possibility of ignition of released hydrogen gas and otherwise mitigates the consequences of hydrogen release. Guidance is given to reactor manufacturers, utility companies, regulatory agencies, and research organizations committed to reducing risk factors and insuring safety of life, equipment, and environment.

## I. INTRODUCTION

The public risk of radioactive exposure from LWRs may be decreased by "improved training for operators on how best to cope with accidents" and by "the upgrading of some reactor safety systems, components, and instruments."[1] It may also be decreased by "wider planning by utilities and state and Federal agencies on how to protect the public in a nuclear emergency,"[1] but this step is not discussed in this paper.

The primary goal of this paper is to present an engineering approach to the complex problem of hydrogen safety in LWRs. Discussion of the accidents that may cause hydrogen generation in a reactor core is outside the scope of our presentation. Our starting point is the presence of a large amount of hydrogen in the reactor vessel. We proceed from this point to develop a safety strategy that eliminates or minimizes the hydrogen hazard in an LWR plant at the design stage and identifies and reduces the hazard in existing plants. Part of our approach makes use of the existing practice in loss prevention in the process industries. A background for a proposal on a safety strategy will be developed first. This will be followed by specific lines of defense for hydrogen safety in operating and future LWRs.

## II. BACKGROUND

An understanding of what happened in the TMI-2 incident should help us improve reactor safety. The combustion of hydrogen in the TMI-2 containment building occurred after a large release of hydrogen into the air atmosphere of the building. The details of the hydrogen transport, ignition, and burn in the containment atmosphere are being investigated.[2] However, hydrogen combustion is a complex phenomenon. Let us start with a gaseous $H_2$-$O_2$ system. After ignition, the product $H_2O$ is not produced by a simple reaction like

$$H_2 + \frac{1}{2} O_2 \rightarrow H_2O \ .$$

Instead, there is a sequence of reactions that includes chain branching reactions (CBRs), as shown in Fig. 1 (Ref. 3). The CBRs produce two $H_2O$ molecules and three H-atoms for every H-atom consumed. To calculate the rate of burning of hydrogen, it is necessary to know the rate constants of each of the individual reactions. Presence of $H_2O$ in the gas mixture may not be favorable for the chemical reactions involving $H_2O$ production. In addition, a reduction in available $O_2$ hinders and, eventually, stops propagation of CBRs. Also, nitrogen enters into additional reactions with oxygen, hydroxyl, and hydrogen radicals. The most important of these reactions are[4]

$$\begin{aligned}
O + N_2 &\longrightarrow NO + N, \\
N + O_2 &\longrightarrow NO + O, \\
N_2 + O_2 &\longrightarrow N_2O + O, \\
N_2O + O &\longrightarrow 2NO, \\
N_2 + O_2 &\longrightarrow 2NO, \\
N + OH &\longrightarrow NO + H, \text{ and} \\
N_2 + OH &\longrightarrow N_2O + H.
\end{aligned}$$

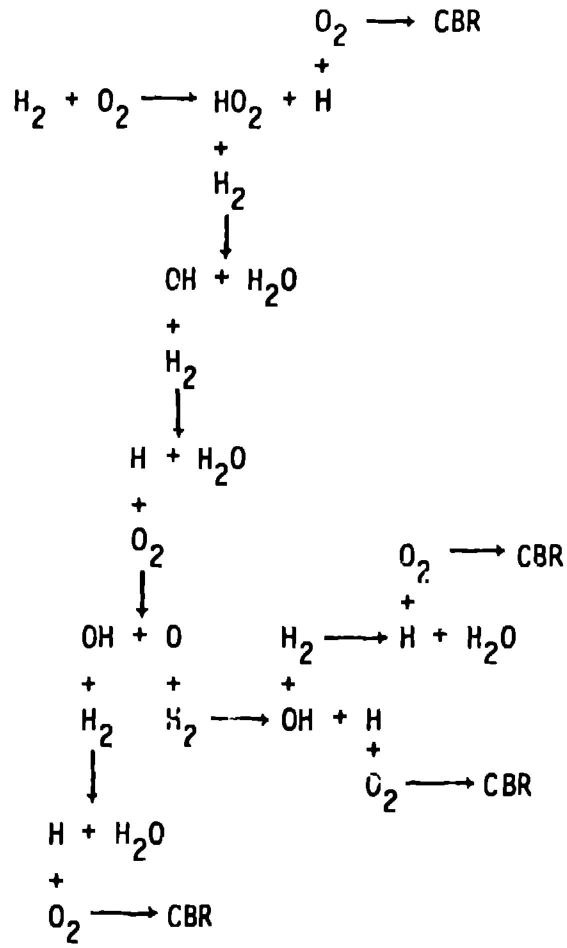


Fig. 1. CBRs in $H_2$-$O_2$ systems.

In simplifying assumptions, the role of $N_2$ is taken to be that of a diluent and its effect is treated as equivalent to a heat loss.

Inhibitors and sensitizers also take part in the chemical kinetics. For example, inhibition by halogens (X for F, Cl, Br, and I) is rapid by the following reaction, which removes the chain carrier H.

$$H + X_2 \longrightarrow HX + X.$$

Also

$$Cl + H_2 \dashrightarrow HCl + H$$

is rapid, although the corresponding reactions with bromine and iodine are slow.

The rate of combustion reactions is usually given in the form

Rate = Constant x Function of Concentrations x Exp(-Constant/T), where T is absolute temperature. Thus, the temperature of the mixture affects the chemical kinetics from ignition to extinction.

In summary, appropriate experiments are needed to provide the following information to better understand the combustion behavior of hydrogen in a mixture with air, $H_2O$, and other possible chemicals.

- Reaction mechanism for interactions between molecules and atoms.
- Rates of reactions.
- Thermodynamic properties and changes in them.
- Initial conditions defined by composition of reactant mixture, temperature, and pressure.
- Final equilibrium state.

Furthermore, studies have shown that, depending on the specific conditions, hydrogen combustion is one of the following three types.

- A phenomenon controlled primarily by chemical kinetics, such as ignition, flame stability, extinction, and quenching of flames (kinetically controlled).
- A phenomenon controlled by diffusion, flow, and other physical mixing processes, such as burning of a gaseous jet, forced turbulence by fans, and presence of inert gases (physically controlled).
- A phenomenon in which both chemical kinetics and physical mixing play important roles, such as premixed hydrogen and air with partial confinement.

In the "kinetically controlled" phenomenon, the reaction rate is slow compared to the rates of heat and chemical species diffusion, which serves to smooth out any spatial nonuniformities. On the other hand, when the reactions are very fast, gradients of species and temperature are established in space. Such gradients cause conduction of heat and diffusion of species towards the regions of lower temperatures and concentrations, respectively. The reactants diffuse away from the flame zone and this poorly mixed combustion is "physically controlled." However, in premixed reacting mixtures, chemical kinetics and diffusion are both important.

Any consideration of hydrogen behavior in closed-air environments has to include the above phenomena. In general, the conditions are governed by the interaction of the supply rate of gases with the rates of chemical reactions. Diffusion and mixing processes between the reactants and removal of the heat and products affect the overall speed.

As a result, the physics to be modeled in a combustion process include conduction, diffusion, viscous action, buoyancy, surface tension, compressibility, kinematics, dynamics, homogeneous reaction, heterogeneous reaction, flame propagation, heat transfer to walls, mass transfer to walls, radiation, mass disappearance, etc. Spalding has identified 125 dimensionless groups that can be encountered in the modeling of these processes appropriate to combustion.[5] Most of the mentioned phenomena play some role in all hydrogen combustion processes. Thus, complete modeling of combustion processes is now practically impossible. However, a partial modeling can be applied to consider only the important influences and disregard the weaker ones. An obvious path is to distinguish between the kinetically controlled and the physically controlled phenomena as discussed above. In other words, instead of attempts at a global hydrogen behavior model, different individual models may be developed to characterize kinetically controlled phenomena such as ignition, flame stability, and extinction; physically controlled phenomena such as burning of jets, forced turbulence by fans, presence of inert gases; or phenomena where other processes play major roles. To the extent that these effects can be separated, better experiments can be carried out for use in phenomenological or consequential understanding of hydrogen behavior.

While scientists study hydrogen combustion, the LWR safety concerns still need to be resolved in the short term. Efforts are under way to control combustion of hydrogen in LWR containments by "suppression of combustion" or "controlled burning."[2] The methods under study for suppression of combustion include inerting, injecting halogens, fogging, and injecting steam. The investigation for controlled burning covers ignitors, catalytic combustors, spark plugs, glow plugs, and open flames. All these efforts assume a hydrogen release into the containment and try to mitigate the consequences of its ignition.

## III. STEPS TOWARDS A SAFETY STRATEGY

In this section, we will start with the TMI-2 incident to indicate the steps that should be taken in the development of a hydrogen safety strategy.

A. Learning Experience from TMI-2.

The sequence of events in the TMI-2 accident that led to combustion of hydrogen is outlined in Fig. 2. Hydrogen was generated in the core by the chemical reaction of zirconium fuel cladding with oxygen and steam. Measurements made at various places in the core help to define the pressure, temperature and other conditions under which the reaction took place. With this information, it is possible to reconstruct the build-up of hydrogen in the core and its transport into the primary coolant system. The latter was influenced by operator actions such as turning on and off pumps and by the coolant system's response to these actions. Then, some of this hydrogen was released to the containment through the stuck-open power-operated relief valve (PORV) and the PORV block valve that the operators had temporarily opened at about 7 h 38 min to depressurize the system. Instruments located in the containment building recorded a pressure spike at 9 h 50 min that is thought to have been caused by the ignition of the hydrogen-air mixture in part of the building. The locations of these instruments can be correlated with calculations to predict the hydrogen concentrations as a function of time in various parts of the containment building. All of these efforts will provide useful benchmarks for our understanding of the various processes.

Our knowledge from this unfortunate event will also be enhanced by a damage evaluation that may help to reconstruct what happened.[6] For example, the overpressures and impulses of the blast wave at different elevations may be estimated from the damage. By working backward, we may obtain a better estimate of the true energy release from the hydrogen. There may be other observations and calculative checks that become obvious after physical examination of the damage in the TMI-2 containment building. This damage analysis may help in further understanding the accident history, as well as testing calculative models.

B. Comparison of Hazard Factors.

Identification of the relative importance of hazard factors in LWR hydrogen safety is important. It is instructive to review, at this point, the American Insurance Association hazard survey of 317 large-loss chemical plant explosions and fires over a 20-yr period.[6,7] Nine hazard factors were identified in the survey, as shown in Table I. Some events involved more than one hazard factor. Almost one-third of these

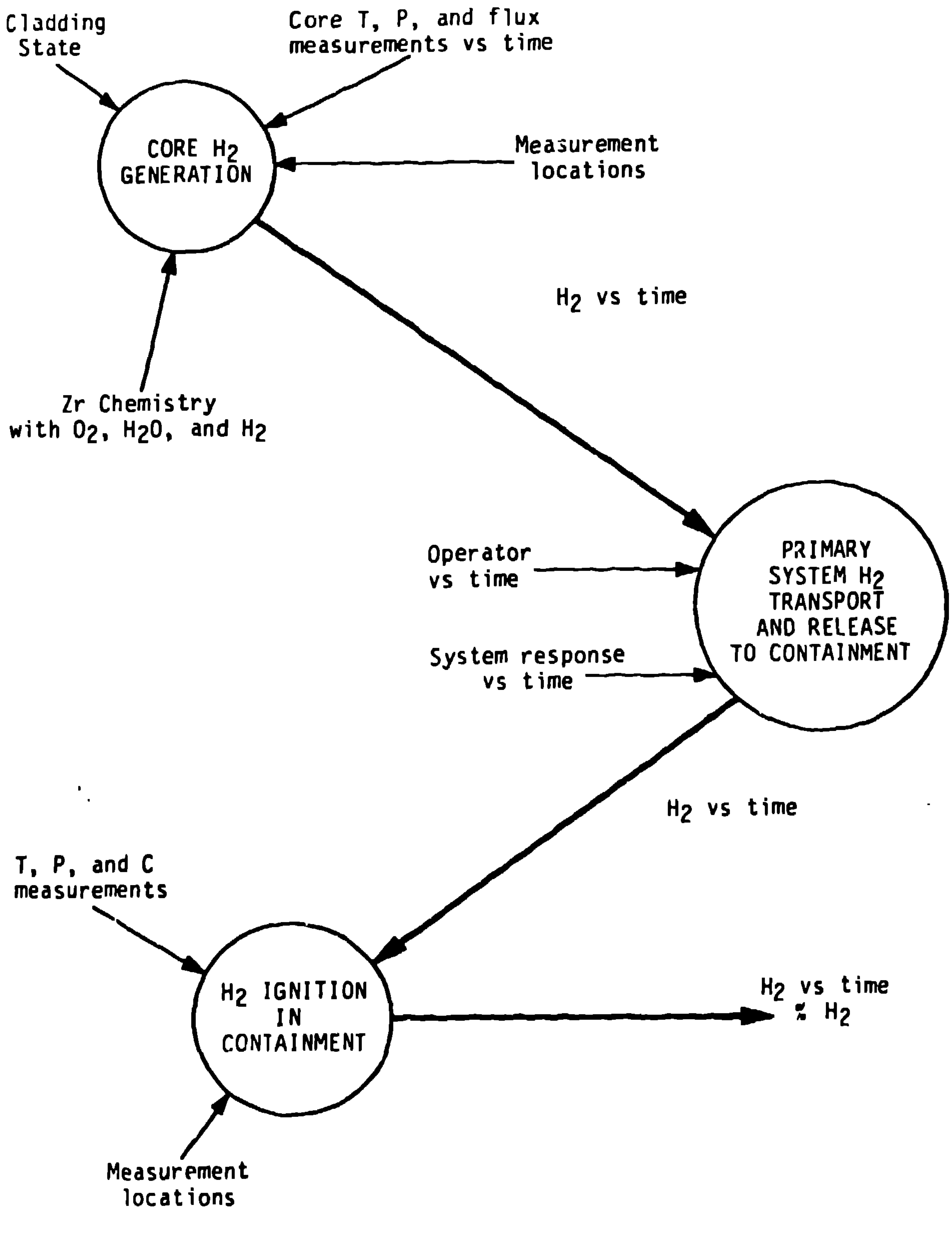


Fig. 2. Sequence of events in TMI-2 hydrogen combustion.

events were caused by equipment failures, one-fifth by inadequate material evaluation, one xth by operational failures, and one-tenth by process problems. These four hazard factors combined accounted for about 80 per cent of all the hazards involved. A loss reduction effort should obviously consider these factors for design, construction, installation, operation, and maintenance of such facilities. In addition, human factors (which were not separated from the others in Table I) are important contributors to hazards. Improper training of operators is recognized as an important hazard factor.

TABLE I

HAZARD SURVEY OF 317 LARGE-LOSS CHEMICAL PLANT EXPLOSIONS AND FIRES[7]

| Hazard Factors | Cases | Per Cent |
|---|---|---|
| Plant Site Problems | 16 | 3.5 |
| Inferior Plant Layout | 9 | 2.0 |
| Poorly Designed Structures | 14 | 3.0 |
| Faulty Material Evaluation | 93 | 20.2 |
| Process Problems | 49 | 10.6 |
| Material Handling Problems | 20 | 4.4 |
| Operational Failures | 79 | 17.2 |
| Equipment Failures | 143 | 31.1 |
| Weak Safety Program | 37 | 8.0 |
| Total | 460[a] | 100.0 |

[a]Greater than 317 because some events involved more than one hazard factor.

Hazard analysis is a valuable tool for collecting all relevant information on a subject and for quantifying as much of it as is reasonably practicable in terms of benefits and drawbacks.[8] A properly executed hydrogen hazard analysis in LWRs should be an aid to the decision makers. However, criteria for acceptable risk should be available beforehand to use in hazard analysis. Once the magnitude and probability of risk are specified quantitatively, hazard analysis may be useful not only to compare alternative routes to the same goal but also to decide whether a situation is acceptable.

C. Expanding the Information Base.

Since the early days of combustion research, the literature has expanded with new data on hydrogen-oxygen systems. Much data are available for LWR applications.[9-13] Also, many regulations, standards, and guidelines have been developed for hydrogen, as summarized by Hord.[14] For example, there are nonmandatory, but industrially accepted, standards for explosive criteria, leak detection, and fire protection. These standards should be useful in improving the hazard profile of LWRs for hydrogen. However, as mentioned previously, we find that incomplete knowledge exists in the following areas relevant to hydrogen combustion phenomena.

- Dispersion of $H_2$ after a release.
- Effects of concentration gradients upon energy release and blast pressure.
- Effect of partial confinement upon transition from deflagration to detonation.
- Effect of size of combustion volume upon transition from deflagration to detonation.
- Combustion of mixtures of gases, including steam.
- Effect of flame suppressants.

An improved information base for the above phenomena would also help to verify analytical tools, as discussed below.

D. Establishing and Verifying Analytical Tools.

Mathematical models are needed for transient, multicomponent, chemically reactive gas flow in confined volumes. Specifically, analytical tools are needed to predict the effects of the following on $H_2$-$O_2$-$N_2$ systems with and without $H_2O$ vapor (including saturation) and with and without additives (inhibitors).

- Confinement in large volumes and in different shapes of volumes.
- Natural convection and buoyancy, including stratification and its effects.
- Partial confinement by structures.
- Simultaneous multiple chemical reactions.

It is desirable to be able to predict experimental results, such as burning velocity and pressure, temperature, and concentration profiles with time, and to extrapolate these to different conditions and confinement situations, including parametric studies for TMI-type cases. Only analytical models thus verified by experimental data can provide the needed tools for analysis of hydrogen behavior in LWR containments.

E. Developing and Qualifying Safety Equipment.

Development of hydrogen safety equipment may be necessary for the "hydrogen isolation" and "hydrogen disposal systems" discussed in the next section. The purpose of this hardware development and qualification is to design, construct, and operate equipment with a desired reliability to burn, combine by chemical reaction, or store hydrogen existing in a system. Off-the-shelf equipment, such as glow plugs, may need proof testing for performance effectiveness. In addition, new or additional instrumentation may be required for detection, monitoring, and safety functions.

F. Establishing Additional Procedures.

Depending on the changes in safety strategy for LWRs, new and additional procedures may be needed in the following areas.

- Operator training for safety equipment and related instrumentation.
- Plant operational procedures, including hydrogen safety systems (such as inerting).
- System interface procedures for new or revised safety functions.
- Plant maintenance procedures, including those for fire protection.

A human operator task analysis should be helpful in identifying workable procedures.

## IV. SAFETY STRATEGY FOR LWRs

A safety strategy can be developed in sensible steps to achieve a desired level of LWR safety. The TMI-2 experience may be used as a benchmark for hydrogen hazard assessment, as discussed in the previous section. Equipment or procedures cannot be evaluated properly without an overall philosophy for safety.

Six levels of defense against hydrogen damage are recommended for consideration and evaluation before implementing them in existing or future LWRs. These levels of the safety hierarchy are as follows.

1. Hazard Analysis of Facility. To minimize the possibility of ignition of flammable gas, the greatest effort should go into the design, installation, and maintenance of any facility.[15] A hazard analysis can be used to approximate the consequences of a major gas

leakage in the cold region of the plant, with subsequent ignition in the hot region of the plant. Study of the plant layout can determine the low-hazard and high-hazard regions for hydrogen safety. Depending on the safety criteria, these areas should receive different individual treatment during the design phase. For example, some equipment may need to be explosion proof; electrical equipment rooms may be located in low-hazard areas; and ventilation intake ports may be at high elevations rather than at ground level. In addition, the margin of safety can be increased by preventing the accumulation of flammable concentration inside critical areas in LWRs. This level of safety review may be most beneficial for future LWRs because it allows changes or modifications at an early phase. However, design and equipment modifications are possible for existing LWRs.

2. Control in the Vessel. The first place to solve the hydrogen problem, once it is generated, is at the source, that is, in the reactor vessel. Although the hydrogen is in a mixture with steam and some oxygen, the volume of gas is much smaller than would be the case after a release into the containment. By venting hydrogen from the top of the reactor vessel, it can be directed to a hydrogen isolation system. This system can use either burning, combiner reactions, or storage processes to isolate the hydrogen. Another way of isolating hydrogen may be by introducing chemicals into the reactor vessel to react with the free hydrogen and/or oxygen.
3. Control in the Primary System. If failure or inadequate operation of the vessel's isolation system should occur, the next place to isolate the hydrogen is in the primary system. A side stream from the hot leg may be used to isolate the hydrogen in a hydrogen disposal system. Just as in the reactor vessel, hydrogen would be in a mixture with steam and some oxygen.
4. Vent from the Primary System. This level provides venting from some high location in the primary system equipment, for example, the pressurizer. Here a porous plug could possibly be used to regulate the flow, followed by combustion or chemical combination of hydrogen.
5. Mitigate Consequences of Release to the Containment. This level is intended to mitigate the hydrogen hazard in the containment. The

equipment and combustible items need to be protected. The high hazard regions identified in the facility hazard analysis may be fortified with hydrogen safety equipment, such as glow plugs and recombiners, or inerting systems and steam curtains. Halon may also be used to prevent combustion.[16] Here one would have to deal with a hydrogen mixture with air, $H_2O$, and possibly other chemicals. Because of dilution with air the gas volumes to be handled are large. An understanding not only of the hydrogen behavior in the containment environment but also of the operational capabilities of safety equipment is necessary to assure loss prevention for equipment. New operational and maintenance procedures may be developed for fire protection.

6. Containment Building. The containment building wall is the last level of defense for protection of life and environment. Loss prevention in plant equipment may also require the implementation of a number of the lower level defenses before this level.

Each of these six levels of defense should be evaluated individually and in combination for effectiveness before deciding which to implement for LWR hydrogen safety. Some of them may not buy much in prevention or may interfere with the existing safety systems. Hence, a systems approach should be applied to prevent interface problems with the other systems. A strategy for containment designs may emerge from such an analysis and evaluation.[17]

## CONCLUSIONS

We believe that the TMI-2 data and the corresponding observational and calculative analyses, a hydrogen hazard assessment of LWRs, and a definition of acceptable safety performance should be used to develop criteria for hydrogen safety in existing and future LWRs. In general, these criteria may lead to

- reactor and plant system modifications;
- new equipment and instrumentation, including hydrogen isolation and disposal systems; and
- new procedures and different system interface requirements.

We have identified the following six steps towards a hydrogen safety strategy.

- Learning from TMI-2.
- Comparison of hazard factors in LWRs.
- Expanding the information base.
- Establishing and verifying analytical tools.
- Developing and qualifying safety equipment.
- Establishing workable procedural methods.

These steps were used to develop a hydrogen safety strategy in six levels of defense. We believe that these levels of defense against the hydrogen hazard should be considered and evaluated before deciding what should be implemented. Also, differences in reactor plant designs may require an individual treatment rather than a generic solution to the hydrogen hazard problem. The outcome should be a reduced level of risk to the public because of the TMI-2 incident.

The Canadian Atomic Energy Control Board's bulletin on "Risk of Energy Production" states that "Risk to human health does not start when an electricity generating plant or solar plant puts out energy. It starts when an engineer first puts pencil to paper, continues as materials are acquired, fabricated and installed, and terminates only when the installation is dismantled at the end of its useful life.... In other words, all risks must be included for a fair evaluation, not just the risk which is most obvious."[8]

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