



UNITED STATES
NUCLEAR REGULATORY COMMISSION
WASHINGTON, D. C. 20555

~~SECRET~~ TERA

JUL 24 1979

Docket Nos. 50-549
50-596
and 50-597

Dr. Peter D. G. Brown
Chairman of the Board
Nuclear Opponents, Inc.
P. O. Box 666
New Paltz, NY 12561

Dear Dr. Brown:

In accordance with your request, we have reviewed the paper by Dr. David M. Pisello, "The Zirconium Connection," transmitted by your letter dated May 1, 1979. Dr. Pisello's paper contains a great deal of incorrect information, of which the following are examples:

1. The charge that NRC concealed the nature of the Zircaloy-water reaction is false. This reaction was discussed extensively in the public hearing on Emergency Core Cooling Systems, and is so routinely analyzed that it is covered explicitly by a regulation (10 CFR 50, Appendix K, paragraph I.A.5).
2. The charge that NRC concealed the source of hydrogen in TMI-2 and only admitted it privately is false. The Zircaloy cladding reaction with water was described as the major source of hydrogen in NUREG-0557, "Evaluation of Long-Term Post-Accident Core Cooling of Three Mile Island, Unit 2," May 1979 and in other public references.
3. The decay heat was not "only about one fifth of what it would be in a mature core." Because the decay heat of concern (i.e., within a few hours after shutdown) depends primarily on short-lived fission products, the decay heat was about 95% that of a mature core at the time of major core damage.
4. The oxide film on zirconium is not transparent to hydrogen, but is nearly impermeable to hydrogen.
5. Plutonium dioxide formed in nuclear fuel is a ceramic; it is not volatile.

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Dr. E. A. Gulbransen, whose work is referenced by Dr. Pisello, has expressed his concerns about the use of zirconium alloys as reactor fuel cladding. We have recently prepared a response to those concerns and are enclosing a copy of that response for your information. In summary, our evaluation of Dr. Pisello's paper indicates that it is without substantial technical merit, and that his allegation of a major design flaw due to the use of zirconium alloys as cladding material is unsupported and unwarranted.

Sincerely

Original signed by
E. G. Case



Harold R. Denton, Director
Office of Nuclear Reactor Regulation

Enclosure:
Gulbransen Response

ENCLOSURE

Evaluation of Prof. Gulbransen's Letter on Zirconium Hydride in TMI-2

Senator Heinz transmitted a letter from Prof. E. A. Gulbransen (U. of Pittsburgh) expressing concern about the formation of zirconium hydride in the TMI-2 core. A similar letter (Enclosure 1) from Prof. W. E. Wallace (a colleague of Gulbransen at U. of Pittsburgh) also reached the NRC, and a response to that letter was prepared earlier (Enclosure 2). Both Gulbransen and Wallace believe that the hydrogen in the TMI-2 "bubble" will react (or has reacted) with the remaining unoxidized Zircaloy to form large quantities of zirconium hydride. Wallace believes that the hydride will be finely divided and lying at the bottom of the reactor, and that this hydride presents a grave explosion hazard if it is exposed to dry air (powdered zirconium hydride is pyrophoric). Gulbransen, on the other hand, believes that the zirconium hydride will react with water to produce more free hydrogen, and that this cycle will repeat until all of the Zircaloy cladding is oxidized and the primary system contains large quantities of free hydrogen.

Both of these concerns are based on Gulbransen's early work on hydrogen reaction with preoxidized zirconium. Gulbransen's experiments were conducted in a vacuum furnace into which hydrogen was admitted. The oxide film was found to be nearly impermeable to hydrogen except at specimen edges where localized attack occurred on chance patches of fresh zirconium. The hydride would then spall exposing more fresh zirconium such that the reaction continued.

Zircaloy cladding in a reactor, however, is in an oxidizing environment (water and steam) such that the fresh edges in Gulbransen's experiments would not be present. If by chance any fresh metal were exposed in the preoxidized cladding, those places would quickly oxidize and seal off the transport of hydrogen.

It is clear that hydriding is not a problem during normal reactor operation even though PWRs are operated with a hydrogen overpressure and cladding temperatures are amply high (about 600°F) for hydriding. Some fuel rods have been kept in reactors for test purposes as long as 15 years without showing evidence of excessive hydriding.

Fuel rods that have experienced abnormal hydriding also demonstrate the effectiveness of an oxidizing atmosphere in shutting off the hydriding process. Earlier industry problems with internal hydriding occurred only after the oxidizing environment had been removed, i.e., the enclosed moisture had been removed by oxidation with zirconium freeing hydrogen. Although hydriding in those cases originated on the cladding inside surface, the hydride phase extended through the wall thickness and was visible on the outside surface as a blister or "sunburst." Those blisters do not spall or continue to hydride significantly on the outside surface even at operating temperatures.

It remains, therefore, to examine the conditions at TMI-2 that were significantly different than our common experience with Zircaloy. These conditions were present during (a) the period of the high-temperature excursion, and (b) the later low-temperature period during which the bubble containing hydrogen was present. The questions to address are (a) whether large amounts of hydrogen are absorbed during a LOCA-like temperature and oxidation transients, and (b) whether hydrogen permeation through the oxide film is significant at relatively low temperatures but high hydrogen pressures.

Hydrogen absorption during temperature transients in the presence of steam has been studied by Kawasaki et al.* They showed that significant hydrogen absorption occurs only when the hydrogen-to-steam ratio is greater than about 0.2. During a LOCA-like temperature transient, this condition is satisfied locally (on the inside of a freshly burst tube near the burst location) resulting in local concentrations of hydrogen only as high as about 3,000 parts per million (i.e., 0.3%). This concentration does not

*Recent results from U.S.-Japanese information exchange. See memorandum dated December 20, 1978 from S. Levine to H. R. Denton (Public Document Room accession number 7902270150).

constitute the severe hydriding problem of concern to Gulbransen, and it is so low that hydrogen pickup was overlooked in many early LOCA-simulation tests.

The rate of hydriding in oxidized zirconium is controlled by the rate at which hydrogen can permeate the oxide film and reach the metal. The effect of hydrogen pressure on the hydrogen permeability of oxide films on zirconium at relatively low temperatures has been studied by Smith (Journal of Nuclear Materials, Vol. 18, p. 323, 1966). In principle, the rate of permeation (and hence the rate of hydriding) is a function of the fraction of available surface (oxide) sites that contain hydrogen, and this in turn depends on the hydrogen overpressure. When the oxide surface is saturated with hydrogen, further increases in the availability of hydrogen, i.e., the hydrogen overpressure, should have no effect. Smith confirmed this effect experimentally over the pressure range of 1 to 860 mm Hg of hydrogen and expressed the pressure effect by the function $bP/(1+bP)$, where P is the pressure and b is a constant. Unfortunately, near the high pressure end of the pressure range in Smith's experiment, results were erratic and the permeation rates were all abnormally high. Therefore, we cannot say with absolute confidence that there is no high pressure effect on permeation, but we believe that to be the case.

The exact time, temperature, and hydrogen pressure conditions are unknown for the TMI-2 accident so we cannot be sure that the TMI-2 conditions were present in the above references. Nevertheless, it seems most likely that the oxide layer that is formed on all Zircaloy cladding during fabrication (by autoclaving) and the additional oxide formed on the damaged TMI-2 cladding will prevent large scale hydrogen absorption and hydriding in the TMI-2 core. We have seen no evidence to suggest the contrary.

Professor Gulbransen also stated that circulating cooling water would react with zirconium hydride forming more zirconium dioxide. This reaction would continue (if zirconium hydride were present) only to the extent of forming an oxide layer on the zirconium hydride. This oxide coating would then inhibit further oxide formation just as it does on the metal.

In his letter, Professor Gulbransen expressed an additional concern about the possible effect of hydrogen on the reactor vessel and piping at TMI-2. At high temperature, hydrogen has the potential for combining with carbon in steels to form methane, which results in internal stresses that can cause cracking. This is sometimes referred to as "hydrogen embrittlement," although a more appropriate term might be hydrogen-induced decarburization. Other types of hydrogen embrittlement are found in some high-strength steels, but it is not encountered significantly in reactor pressure-vessel steels.

Two circumstances should prevent hydrogen-induced decarburization from being significant in the pressure vessel and piping steels at TMI-2. (1) The pressure vessel and large pipes in the primary system are lined with stainless steel, which acts as an effective barrier to hydrogen and will prevent its contact with the higher-strength alloys. (2) The effect of hydrogen on steels of the type used in pressure vessels has been found to be unimportant for the combination of pressure and temperature experienced in TMI-2 (even if stainless steel lining were not present). Enclosure 3 discusses this topic and shows that these steels can be used indefinitely at temperatures up to 700°F at a pressure of 1000 psi of hydrogen. Except for short periods during the first day of the accident--periods of time that would not meet incubation requirements for this phenomenon--, total system pressure was 1000 psi or less (hydrogen partial pressure would, therefore, be smaller) and system temperatures were generally below 600°F. It therefore seems unlikely that hydrogen embrittlement would have occurred in the primary system. In any event, TMI-2 inspections for damage would be made prior to operating that reactor again.

In conclusion, there is strong evidence that the type of cladding hydriding feared by Gulbransen and Wallace will not occur because of the continued formation of a protective oxide layer. Even if zirconium hydride were formed, there is little danger that it could ignite because the damaged TMI-2 core will not come into contact with dry air until after underwater examinations could determine the presence of significant zirconium hydride. It will be an easy matter at the time of examination to see if Gulbransen's prediction of a fully reacted core is valid; we think it is not. The explosion risk of free hydrogen in the primary system is also small.

Hydrogen in the primary system is being regularly monitored and found to be very low in concentration--so low, in fact, that hydrogen is being added to adjust the pH. The effect of hydrogen on piping and vessel materials during the period of the TMI-2 bubble's existence should also be insignificant since that exposure to hydrogen was within temperature and pressure limits for which no damage is expected.

May 24, 1979

SUBJECT: Zirconium Hydride

TO: Richard Vollmer, Asst. Director, Systems and Projects, NRC
 Robert Arnold, Metropolitan Edison Company, Reading, Pa.
 Thomas Gerusky, Director, Bureau of Radiation Protection, DER

FROM: Clifford L. Jones, Secretary *CLJ*
 Department of Environmental Resources

I am enclosing a letter from Dr. W. E. Wallace who is a Distinguished Service Professor at the University of Pittsburgh with a speciality in chemistry.

Dr. Wallace and one of his associates, Dr. Earl Gulbransen, feel that there may be problems with zirconium hydride at the reactor as it is cooled. Dr. Gulbransen has offered his time without cost as a consultant on this problem.

These people are reliable individuals who are concerned and have knowledge and experience to offer. I would appreciate it if you would be in touch with Dr. Earl Gulbransen, his telephone numbers are on the attached letter and see if he can be of help. Thank you.

Att:
 cc: Governor Dick Thornburgh
 Lt. Governor William Scranton, III
 William Middendorf



University of Pittsburgh

FACULTY OF ARTS AND SCIENCES
Department of Chemistry

May 18, 1979

Mr. Clifford Jones, Secretary
Department of Environmental Resources
Commonwealth of Pennsylvania
Harrisburg, PA 17120

RE: Chemical Explosive Hazard at Three Mile Island

Dear Cliff:

This is to put in writing the substance of my comments to you via telephone a few moments ago.

I am reasonably certain that the hydrogen produced at the Three Mile Island accident is still there and represents a grave explosion hazard if improperly handled. My authority for this is Dr. Earl Gulbransen, Research Professor of Metallurgy and Materials Engineering and of Chemistry, in this University.

Clearly the hydrogen was formed by reaction of the zircalloy cladding and water in the overheated reactor. To the best of my knowledge its disappearance, i.e. of the hydrogen bubble, is generally regarded as a mystery. Dr. Gulbransen asserts, and I agree with him, that it disappeared as a gas by reaction with zircalloy to form zirconium hydride as the reactor cooled (thermodynamic measurements of Dr. Gulbransen fully support this idea). He furthermore holds the opinion that some tons of this exceedingly hazardous hydride are lying at the bottom of the reactor where it is at present covered over with water. As long as this hydride is covered, it presents no hazard; but there is a distinct possibility of an explosion when this finely divided pyrophoric mass comes in contact with oxygen in the air. This presents a special hazard at the time of the drainage of the reactor.

My strong recommendation is that Dr. Gulbransen be used as a consultant in regard to the clean-up operation at this reactor site. He had anticipated this "hydrogen problem" some years ago. My credentials in the hydrogen area are strong also, and I would be willing to give what help I can. However, I will be in Japan from May 19-June 3, inclusive. Dr. Gulbransen's address and phone numbers are as follows:

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Dr. Clifford Jones, Esq. ary
May 18, 1979
Page 2

Home: Dr. Earl Gulbransen
63 Hathaway Ct.
Pittsburgh, PA 15235

Phone: 412-793-0141 *home*
412-624-5312 C.G.

Office: Dept. of Metallurgy and Materials Engineering
University of Pittsburgh
Pittsburgh, PA 15260

Phone: 412-624-5312

If you need me, I can be reached in Japan as follows: May 21-24, Hakone Prince Hotel, Hakone; May 25-27, Holiday Inn, Kyoto, Japan; May 28, 29 in Tokyo with Jim Conté -- Telephone: 03-387-4758; May 30, 31, June 1, Kauai Surf Hotel, Kauai; June 2 with my son, Wallace in Seattle -- Telephone Contact: 206-631-5336. I will be back at my office (412-624-5004) on June 4.

Be sure that someone alive to the chemistry of the problem is involved in the clean-up effort. I know you will take steps which are appropriate in following up on this letter.

Sincerely yours,

W. E.
W. E. Wallace
Distinguished Service Professor

WEN/ams

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UNITED STATES
NUCLEAR REGULATORY COMMISSION
WASHINGTON, D. C. 20555

JUN 06 1979

MEMORANDUM FOR: Paul S. Check, Chief, Reactor Safety Branch, DOR
THRU: Carl H. Berlinger, Section Leader, Reactor Safety Branch, DOR
FROM: Kris I. Parczewski, Reactor Safety Branch, DOR
SUBJECT: FORMATION OF ZIRCONIUM HYDRIDES IN THE THREE MILE ISLAND-2
INCIDENT

CJB

Introduction

The Secretary of the Department of Environmental Resources, Commonwealth of Pennsylvania transmitted to us a letter from Professor W. E. Wallace of the University of Pittsburgh in which he draws attention to the fact that during the TMI-2 accident large amount of generated hydrogen may have caused formation of zirconium hydrides which, if not handled properly, can under certain circumstances cause a violent reaction. Prof. Wallace quoted the work of Professor E. Gulbransen, also from the University of Pittsburgh, who for the last 25 years was studying the kinetics of formation and decomposition of zirconium hydrides.

The purpose of this memo is to evaluate, in light of the presently available information, the concerns brought by Prof. Wallace.

Available Information

The information used in evaluating the problem of zirconium hydrides came from the following sources:

- (1) Telephone conversation with Prof. Gulbransen (06/04/79).
- (2) Conversations with several members of the NRC Staff (F. D. Coffman, M. L. Picklesimer, D. A. Powers).
- (3) "The Metallurgy of Zirconium," by B. Lustman and F. Kerze, Jr., Mc Graw-Hill Book Company, Inc., 1955.
- (4) "The Metallurgy of Zirconium," by D. L. Douglass, IAEA, Vienna, 1971.
- (5) "The Encyclopedia of the Chemical Elements," by C. A. Hampel, Reinhold Book Corporation, 1968.
- (6) "Dangerous Properties of Industrial Materials," by N. I. Sax, Van Nostrand Reinhold Company, 1975.

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JUN 06 1979

Evaluation of the Problem

Prof. Gulbransen (Source 1) had indicated that when Zr comes in contact with hydrogen at certain pressures two types of zirconium hydride are formed: Zr H_{1.4} and Zr H_{1.9}. At about 500°C the equilibrium hydrogen pressures for these compounds are few hundredths of mm Hg and few mm Hg, respectively. These hydrides are formed despite the existence of protective ZrO₂ because, according to Prof. Gulbransen, ZrO₂ cannot stop completely penetration of hydrogen into metallic Zr. This is a controversial point since in the opinion of other people (Source 2) ZrO₂ could completely prevent hydrogen from coming in contact with metallic Zr. The information from the literature (Sources 2 and 3) also confirmed the view that ZrO₂ would very significantly limit hydrogen penetration.

Prof. Gulbransen pointed out that Zirconium hydride formed on Zr surfaces may spall off forming a highly divided mass at the bottom of the reactor vessel. This point was also challenged by other people (Source 2) who did not believe that Zr hydride could ever assume a highly divided form.

According to Prof. Gulbransen the presence of zirconium hydride in the reactor vessel in TMI-2 could cause two problems:

- (1) In contact with water at lower pressures hydrogen gas can be released. Although the rate of release would be slow the existence of this source of hydrogen should be taken into consideration.
- (2) Zirconium hydride in powdery form is pyrophoric and when exposed to air may ignite and produce violent reaction. The information obtained from other sources (Source 6) shows that the auto-ignition temperature of Zirconium hydride is 270°C in air. It is, however, very much dependent on the physical form of the hydride.

As a remedy Prof. Gulbransen has suggested a method for decomposing zirconium hydrides by circulating hydrogen free water at low pressure and preferably containing some oxidizing agent (e.g. dissolved air). The rate of decomposition will be slow because of a slow rate of reaction and it would take a long time to decompose all hydrides.

In order to determine the maximum amount of zirconium hydride which could theoretically be formed during the accident it was assumed that 30% of Zr in the core reacted with steam or water and that 30% of the hydrogen generated in this reaction formed hydrogen hydride. With these assumptions about 2500 lb of zirconium hydride would be formed in the reactor vessel during the accident. It should be realized however, that this is an upper theoretical limit and it is most unlikely that such large amount of zirconium hydride would ever be produced.

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Paul S. Check

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Conclusion

The existing information on formation and behavior of zirconium hydride are somewhat controversial, however, because of the possibility of existence of this hazardous material in the reactor vessel the following precautions are recommended:

- (1) To monitor the presence of hydrogen in the primary coolant in order to establish if the decomposition of zirconium hydride takes place.
- (2) When opening the reactor vessel for cleaning assure that the debris at the bottom of the vessel are not exposed to the oxidizing environment (e.g. dry air).

K. I. Parczewski

Kris I. Parczewski
Reactor Safety Branch
Division of Operating Reactors

cc: C. Berlinger
F. Coffman
S. Weiss
R. Vollmer

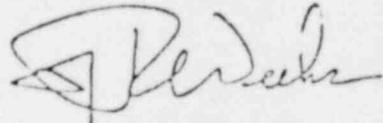
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BROOKHAVEN NATIONAL LABORATORY

MEMORANDUM

DATE: April 4, 1979

TO: W.Y. Kato

FROM: John R. Weeks 

SUBJECT: Discussions Relative to the
Three Mile Island Incident

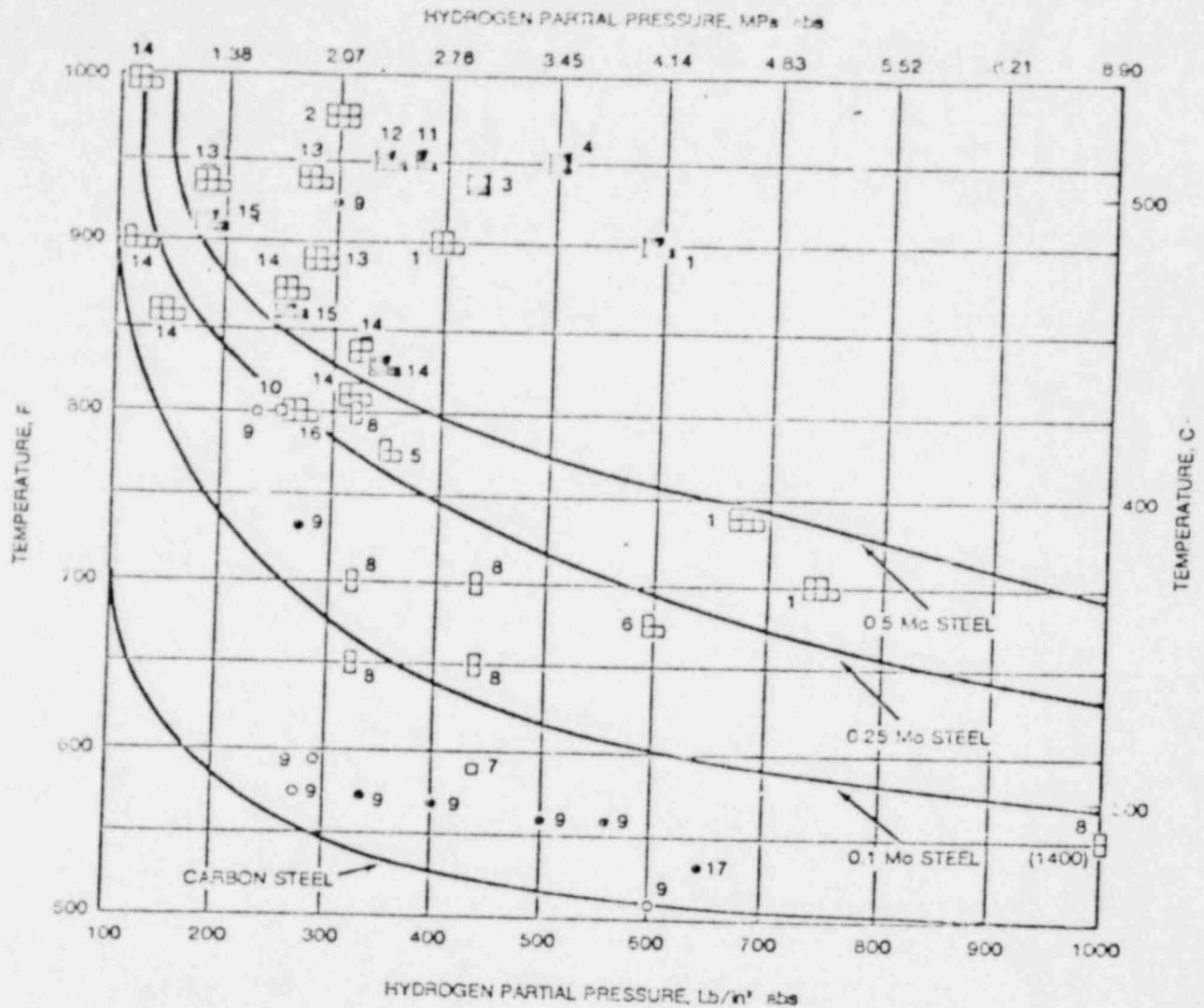
1. Hydrogen in Containment

Walter Butler of NRC asked me to estimate being possible the build-up of hydrogen in the containment by radiolysis of water in a high γ field. I in turn discussed it with Dr. Harold Schwarz of the BNL Chemistry Department. His rough guess was that the hydrogen may build-up to several percent which should be approaching the ignition point. The higher the temperature (above 100°C), however, the greater would be the recombination rate and the less the build-up of hydrogen.

2. Discussions Concerning the Hydrogen Bubble in the Reactor Vessel

Warren Hazelton asked me what information I had on the thermodynamics and kinetics of the reaction of hydrogen at a high temperature and pressure inside the reactor vessel on the possible decarburization of and methane formation in the vessel material. I discussed this subject with David Gurinsky and J. Chow of BNL, M. Gensamer, Professor Emeritus at Columbia and A. Ciuffreda of Exxon Research. The stainless steel cladding on the inner surface of the vessel would be a partial barrier to hydrogen provided it were intact. There is enough of a chance of a flaw in this cladding, however, that no credit should be taken for it in estimating the performance of the reactor vessel material. The reactor vessel is made of a pressure vessel steel (ASTM A-533-B) which contains 1% Mn, 0.5% Ni and approximately 0.5% Mo. The oil industry is continuously concerned about hydrogen induced decarburization of steels in their refinery equipment. They have prepared a graph stating the safe temperature and pressure for steels (Nelson Diagram) in the American Petroleum Institute report API-941, which was most recently modified in 1977. A steel of the composition used in the Three Mile Island vessel should be safe from decarburization by 1000 psi of hydrogen at temperatures up to 700°F for indefinite use. Exceeding this temperature or pressure for short periods would not cause serious damage as there is a definite incubation time, of a matter of several days, before problems begin to develop. Mo appears to be even more effective than Cr in retarding this decarburization although the reasons are not clear. The same steel without the Mo would only be safe up to 500°F at 1000 psi of hydrogen. I think the upper part of the reactor vessel should be carefully checked for any possible damage from decarburization prior to its return to service. A copy of the curve showing this relationship as revised in 1977 is appended to this memorandum.

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References

References	Analysis		Mo Equivalent
	CR	Mo V	
1. Shell Oil Co.*	0.50		0.50
2. Weld Deposits, D.J. Bergman*	0.79	0.39	0.59
3. Weld Deposits, D.J. Bergman*	0.80	0.15	0.35
4. Weld Deposits, D.J. Bergman*	0.50	0.25	0.37
5. Continental Oil Co.*		0.25	0.25
6. Standard Oil Co. of California*		0.27	0.27
7. Standard Oil Co. of California*	0.05	0.06 0.08	
8. A.O. Smith Corp.*		0.13-0.18	0.11
9. Shell Development Co. Drawing No. VT 659-2,			
10. AMOCO Oil Co.*	0.04		0.01
11. R.W. Manuel, <i>Corrosion</i> , 17 [9], pp. 103-4, Sept., 1961	0.27	0.15	0.22
12. The Standard Oil Co. of Ohio*	0.11	0.43	0.50
13. Exxon Corp.*			
14. Union Oil of California*			
15. Amoco Oil Co.*			
16. Standard Oil Co. of California*			
17. Gulf Oil Corp.*			

LEGEND

EQUIVALENT MOLYBDENUM CONTENT	MOLYBDENUM CONTENT						
	0.01-0.10	0.11-0.20	0.21-0.30	0.31-0.40	0.41-0.50	0.51-0.60	
SATISFACTORY	○	□	▣	▤	▥	▦	
HYDROGEN ATTACK	●	■	▩	▪	▫	▬	

NOTES:

- Mo has four times the resistance of Cr to H₂ attack.
- Mo is equivalent to V, Ti, or Nb up to 0.1 percent.
- Si, Ni, Cu, P, and S do not increase resistance.

From API-941, 2nd Edition, 1977.

*Private communication to Subcommittee on Corrosion

Figure 2—Operating Limits for Steels in Hydrogen Service Showing Effect of Trace Alloying Elements

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April 4, 1979

Hazelton also asked whether radiolysis of the water within the vessel could add oxygen to the hydrogen gas bubble. In my opinion, it should not. Radiolysis of water proceeds by a complex chain reaction and can be prevented even by a small overpressure of hydrogen in an operating PWR. The high hydrogen pressures over the coolant at Three Mile Island should totally prevent oxygen formation. In fact, Harold Schwarz stated it may be feasible to remove the hydrogen by simply adding oxygen slowly to the coolant; this could, admittedly, be risky. I think we should be very careful not to use chemicals such as sulfate or sulfur bearing compounds to react with the hydrogen since these can be reduced by the excess hydrogen to sulfides which are very harmful to a number of the materials in the system, especially the Inconel steam generator tubes. It might complicate the return of the unit to service. I recommended that a nitrate (such as potassium nitrate) be used if one wishes to go by this route. However, I think the best means of hydrogen removal would be through venting it from the primary coolant into the containment where it can be recombined with oxygen.

3. Some Crude Calculations of the Amount of Zircaloy that Participated in a Zr-H₂O Reaction During the Incident

I estimate that as much as 3200 lbs. of Zr may have reacted with water to produce the hydrogen bubble, assuming it occupied 750 cu. ft. at 500°F and 1000 psi, as stated by Hazelton. This suggests that over 10% of the Zircaloy cladding in the core was converted to oxide by reaction with the water. Whether or not the remaining Zircaloy could act to remove hydrogen from the water by hydride formation is not clear. However, the hydrogen overpressure during normal PWR operation does not cause significant hydriding of the fuel cladding so that hydrogen removal from the bubble by this mechanism seems unlikely. This hydrogen (10-50cc STP/kg H₂O) amounts to a maximum of 3.24 lb. in the primary coolant (329,200 kg) so clearly, the majority of the hydrogen bubble came from some other source such as Zr-H₂O reactions, if the bubble was as large as described by Hazelton on 3/31/79.

JRW:ob

Distribution

BNL J. Chow
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