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Sent via email

Catherine J.K. Sandoval
Commissioner, California Public Utilities Commission
San Francisco Office (Headquarters)
505 Van Ness Avenue
San Francisco, CA 94102
CJS@cpuc.ca.gov

Subject: Nuclear fuel rod cladding material

Dear Commissioner Sandoval:

This letter is pursuant to the commitment I made to you on July 26, 2011, to get back to you regarding your question of the problem of hydrogen generation during loss-of-coolant accidents in light water reactors and whether any alternative fuel cladding materials might be available to mitigate or eliminate the problem.

The matter required quite a bit of research and the issue has a long and tangled history. But the gist of it is as follows (a list of references is provided at the end of this letter):

- Zircaloy was chosen because of its excellent properties in a number of respects, including low neutron absorption (important for maintaining the chain reaction economically), heat transfer properties, and other mechanical properties needed in a material that would be made in to ten- or twelve-foot long thin tubes containing nuclear fuel. The hydrogen generation vulnerability has long been recognized – at least since the 1970s – well before the 1979 Three Mile Island accident during which there was a contained hydrogen explosion. Notably, Earl Gulbransen, one of the most eminent authorities on the properties of metals and alloys in nuclear power reactor environments, expressed concern in 1975 that there was no back-up to zircaloy in case it proved unsuitable. He pointed to possible embrittlement of zircaloy during operation and to hydrogen evolution due to zirconium-steam reactions during loss of coolant accidents as principal issues.[1]

[1] Gulbransen 1975. Dr. Gulbransen's note drew a sharp response from N.J. Palladino, a professor of nuclear engineering, but this response only challenged Gulbransen on operational suitability of zircaloy.

- Stainless steel cladding has been recognized as an alternative and has been used in five U.S. light water reactors: Haddam Neck (also known as Connecticut Yankee), Indian Point 1, LaCrosse, San Onofre 1, and Yankee Rowe.[2] Stainless steel reduces but does not eliminate the problem of hydrogen generation, since a similar chemical reaction with steam occurs as with zircaloy. The use of stainless steel cladding was stopped in the 1990s after fuel cladding damage was discovered at the Haddam Neck plant; similar damage does not seem to have occurred at San Onofre 1.[3]
- There seems to be no readily available drop-in substitute for zircaloy at the present time. It appears that silicon carbide would immensely reduce the problem of hydrogen generation (by one or two orders of magnitude).[4] But it has the disadvantages of being brittle and of having lower thermal conductivity. Extensive testing would likely be required for safety and operational reasons before it could be used as a replacement for zircaloy in light water reactors. For instance, this is indicated by computer modeling exercise that examined the use of silicon carbide fuel cladding.[5]

There appears to be no urgent, serious push to greatly reduce (or eliminate) the problem of hydrogen generation due to steam-cladding reactions to the point where the maximum amounts of hydrogen involved would be too small to create catastrophic explosions even in worst case accidents. The NRC Fukushima Task Force apparently decided not to consider this issue. I asked about it, rather insistently, at an NRC public meeting about the report, but did not get a clear response, other than it was not considered and that in any case it involved all 104 U.S. operating reactors.[6] I would have thought that this simple fact would have been an important reason to consider the question and put in squarely in the middle of long-term safety considerations. But the NRC Task Force seems to have decided otherwise.

In view of the central role of the zirconium-steam exothermic reaction in the Fukushima accident, it would be prudent for the NRC to recommend a substantial investment in alternative fuel cladding for existing nuclear reactors, even though some of them now have licenses that extend into the 2040s. The NRC appears disinclined at present to do that. Since it affects a substantial majority of states, an initiative on the part of the states to persuade the federal government to take up the zircaloy replacement issue with a high priority might be considered.

Dr. Palladino did not address the issue of hydrogen generation during a loss of coolant accident in his response. See Palladino 1976. Dr. Palladino was later Chairman of the Nuclear Regulatory Commission during a key regulatory period after the TMI accident (July 1, 1981 to June 30, 1986),.

[2] DOE 1994 p. 23 and Table 10 (p. 28)

[3] Rivera and Meyer 1980 p.1

[4] CANES 2011

[5] Carpenter et al. 2007 Chapter 5

[6] NRC 2011 Report and NRC 2011 Briefing pp. 63-68

Yours sincerely,

Arjun Makhijani, Ph.D.
President

cc
California Public Utilities Commission:

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Carpenter et al. 2007	D. Carpenter, K. Ahn, S.P. Kao, P. Hejzlar, and M.S. Kazimi. *Assessment of Silicon Carbide Cladding for High Performance Light Water Reactors*. (MIT-NFC-TR-098) Cambridge, CANES, Nuclear Fuel Cycle Technology and Policy Program, Massachusetts Institute of Technology, November 2007. Link on the Web at http://canes.mit.edu/reports/assessment-silicon-carbide-cladding-high-performance-light-water-reactors.
DOE 1994	U.S. Department of Energy. *Spent Nuclear Fuel Discharges from U.S. Reactors 1992*. (SR/CNEAF/94-01) Washington, DC: DOE, Energy Information Administration, Office of Coal, Nuclear, Electric and Alternate Fuels, Survey Management Division, May 1994. On the Web at http://www.iaea.org/inis/collection/NCLCollectionStore/_Public/25/070/25070038.pdf.
Gulbransen 1975	Earl A. Gulbransen. "Not Safe Enough," *Bulletin of the Atomic Scientists*, June 1975, p. 5. On the Web at http://books.google.com/books?id=JAsAAAAAMBAJ&pg=PA5&lpg=PA5&dq=earl+gulbranson+zirconium&source=bl&ots=L8ZWTPofuz&sig=Mn_X85Dys82e6nI1s8vwUR4t8Mk&hl=en&ei=ZnEoTp-hLIndgQeX6shc&sa=X&oi=book_result&ct=result&resnum=3&ved=0CCAQ6AEwAg#v=onepage&q&f=false. Letter to the editor.

NRC 2011 Report	U.S. Nuclear Regulatory Commission. *Recommendations for Enhancing Reactor Safety in the 21st Century: The Near-Term Task Force Review of Insights from the Fukushima Dai-ichi Accident*. Washington, DC: NRC, July 12, 2011. On the Web at http://pbadupws.nrc.gov/docs/ML1118/ML111861807.pdf.
NRC 2011 Briefing	U.S. Nuclear Regulatory Commission. *Briefing on the Task Force Review of NRC: Processes and Regulations Following the Events in Japan. July 28, 2011, 1:00 P.M., Transcript of Proceedings, Public Meeting, Rockville, Maryland.* (Work Order 58) [Washington, DC: NRC, July 28, 2011]. On the Web at http://www.nrc.gov/japan/20110728.pdf.
Palladino 1976	N.J. Palladino. "Defends Zirconium," *Bulletin of the Atomic Scientist*, March 1976, p. 5. On the Web at http://books.google.com/books?id=XwwAAAAAMBAJ&pg=PA5&lpg=PA5&dq=%22Earl+A.+Gulbransen%22+Bulletin+of+the+atomic+scientists&source=bl&ots=-SZiErULGS&sig=chMOHIO0thbXU-gQc9g0R-ViH2U&hl=en&ei=tqEoTsKBGK6x0AG8r-3iCg&sa=X&oi=book_result&ct=result&resnum=1&ved=0CB8Q6AEwAA#v=onepage&q&f=false. Letter to the editor; a reply to Gulbransen.
Rivera and Meyer 1980	J. E. Rivera and J. E. Meyer. *Stainless Steel Clad for Light Water Reactor* Fuels. (Energy Laboratory Report No. MIT-EL 80-021) Cambridge: EL, Massachusetts Institute of Technology, July 1980. On the Web at http://dspace.mit.edu/bitstream/handle/1721.1/60538/EL_TR_1980_021.pdf?sequence=1.

permitted themselves to sign this document. I blush for them.

William Girdner
Bronxville, N. Y.

Credibility Lost

Considering the many thousands of scientists presently in the employ of large and influential corporations with billions of dollars riding on nuclear contracts, and taking into account the many universities and national laboratories with millions in research funds allocated by the late Atomic Energy Commission, it is indeed revealing that only 32 scientists would lend themselves to promote nuclear power ("No Alternative to Nuclear Power," *Bulletin,* March 1975).

It seems evident that the overwhelming majority of responsible scientists are troubled by the implications of nuclear energy and the resultant plutonium economy. For the 32 scientists to point the finger at nuclear critics and to charge that "the public is given unrealistic assurances that there are easy solutions" is the height of cynicism. This technique of the pot calling the kettle black is exactly the reason why the public has lost faith in nuclear promoters and why the AEC lost its credibility and had to be disbanded.

Nat. H. Sauberman
Great Neck, N.Y.

We Need Restraint

Reading the statement on nuclear power by 32 scientists in the March issue of the *Bulletin,* I remembered Eugene Rabinowitch's farewell article, "Challenges of the Scientific Age," published in September 1973. Rabinowitch had apparently reached the conclusion that the combined energy and pollution problem could be solved only by heavy emphasis on solar energy and the stabilization of population.

Before concluding that there is "no alternative to nuclear power" one should re-read Rabinowitch's article, which includes the following statement:

> It may well be that an ultimate, steady-state, high-technology mass civilization on Earth will have to be based primarily on the utilization of solar energy.

The recent environmental statement by the former Atomic Energy Commission on the proposed Retrievable Surface Storage Facility (RSSF) makes it clear that we shall not know for a period of from 20 to 100 years whether we can store the high level and transplutonium contaminated wastes which will be produced by our burgeoning nuclear power plants without risk of grave planetary contamination.

With due respect to the authors of the statement, it would have been more impressive if the signatures of ecologists, geneticists, demographers, economists, and political scientists had been attached.

This is a period when restraint, not exuberance, should be exercised with respect to nuclear power until we know where we are going.

Anthony Wayne Smith
Washington, D.C.

Not Safe Enough

I read with great concern the statement given by 32 leading scientists on nuclear power, coal and America's energy future.

The major point of the statement is that we should move on to nuclear power since power based on uranium "is an engineered reality for generating electricity today." The scientists also state that "the safety of civilian nuclear power has been under public surveillance without parallel in the history of technology" and that "on any scale the benefits of a clean, expensive and inexhaustible domestic fuel far outweigh the possible risks."

After 25 years of research and development work on the chemical and metallurgical properties of metals and alloys used in nuclear power plants, I have come to the conclusion that the current design and materials cannot give us a safe and well-engineered nuclear power plant. It now appears that there are serious limitations for some of the materials used in nuclear reactors.

The use of zirconium alloys as a cladding material for the hot uranium oxide fuel pellets is a very hazardous design concept since zirconium is one of our most reactive metals chemically. For a safe reactor the cladding material should be relatively inert to water, impurities in the water and to the supporting structure under any possible reaction condition which may occur in a nuclear reactor.

At the operating temperature of nuclear power reactors zirconium cladding alloys react with oxygen in water to form an oxide layer which partially dissolves in the metal embrittling and weakening the metal tubing. Part of the hydrogen formed in the zirconium-water reaction dissolves in the metal and may precipitate as a hydride phase also embrittling and weakening the metal tubing. Recent work reported last summer in France has shown rapid solution of oxygen from the zirconium oxide layer into the metal at the grain boundaries, which could reduce the effectiveness of zirconium alloys as a cladding material. At 400° Celsius the diffusion coefficient for oxygen, D_{GB}, at the grain boundaries in zirconium was about 10^{-11} square centimeters per second compared to a value of 2.1 times 10^{-16} square centimeters per second for the bulk diffusion coefficient D_V.

At temperatures above 1,100° Celsius zirconium reacts rapidly with steam with a large evolution of heat and the formation of free hydrogen, with most metals to form intermetallic compounds and with other metallic oxides to form its own oxide. Once zirconium is heated to 1,100° Celsius, which could occur in loss of coolant accidents, it is difficult to prevent further reaction, failure of the tubing and of the reactor. It is difficult to define the reaction conditions under loss of coolant conditions and good kinetic measurements for the reaction of zirconium with steam at 1,100° Celsius and higher do not exist.

Many of the recent difficulties in the operation of our present nuclear power plants are due to material problems in the reactor, steam generator and turbine. There appears to be no way to overcome the inherent material problems associated with zirconium alloys and the current design of the reactor.

Greater wall thicknesses for the cladding and lower operating temperatures of the fuel may help but the chemical and metallurgical behavior of zirconium alloys cannot be overcome. No backup or alternative design is available if the present design and materials prove unreliable. It is therefore most important to question the statements made by the 32 scientists before it is too late to change.

Earl A. Gulbransen
Department of Metallurgical and
Materials Engineering
University of Pittsburgh

Too Much

Your March issue with the article, "No Alternative to Nuclear Power," shocks me. As scientists, how can you justify this statement? What are your criteria?

Cancel my subscription at once and refund the balance. Scientists and science publishers that can be influenced away from the strict truth are an abomination and worse cheaters than the Nixon administration. You can destroy the future of our children and their progeny till eternity.

Laina Gerrish
Bennington, Vt.

acceptable methods for long-term disposal of such wastes. The terrorists have much simpler ways of carrying out their operations than getting into the nuclear field, although they might use a threat as blackmail. However, having been involved in the reactor safety field for about 25 years, I am satisfied that the major concern for the public should still be reactor safety.

It is sad that Salzman is identified as a representative of Friends of the Earth. Her letter has done no good to the credibility of that organization among knowledgeable people.

C. A. Mawson

Ottawa, Canada

Defends Zirconium

My attention was recently drawn to a letter, "Not Safe Enough," by Earl A. Gulbransen (*Bulletin*, June 1975), regarding the use of zirconium in nuclear reactors. The purpose of this letter is to place before the reader the record of successful performance of zirconium in nuclear reactors.

At the present time I am a professor of nuclear engineering and Dean of the College of Engineering at The Pennsylvania State University. I have had 29 years of experience as an engineer in the nuclear field including extensive responsibility for nuclear reactor design and development work. In that work I was in close contact with the development, testing, and use of Zircalloy at Oak Ridge, Argonne, and Bettis. But to make sure that my response reflects the latest information in this area, before preparing this letter I contacted two of my colleagues at the Westinghouse Electric Corporation who were directly involved in the development and testing and use of zirconium and its alloys.

Zirconium and its alloys have been used as the major cladding and structural material in the reactor cores of water-cooled naval nuclear plants since 1951. Since that time, the operation of hundreds of reactor cores has shown no deterioration of zirconium resulting from interaction with the water environment or the fuel; post-operation destructive examination has revealed reaction with the environment to be miniscule.

Similar experience has been reported with the use of zirconium alloys as cladding for uranium oxide fuel pellets in commercial nuclear reactor plants beginning with the Shippingport, Pa., plant in 1957. In fact an experiment has recently been concluded in which zirconium clad fuel elements were exposed continuously in the Shippingport plant from 1957 to 1974, a calendar exposure 5 to 10 times longer than the design life of commercial fuel elements; the thickness of the oxide layer, the amount of its solution in the metal, the amount of hydride phase formed on fuel elements examined during this period were all small and far below design limits. Zirconium alloys are also being utilized in Canadian power reactors not only for fuel element cladding but also for the primary pressure boundary, with equally favorable experience; the successful operation of water-cooled zirconium core nuclear power plants in Europe and Asia has also failed to support Gulbransen's fears.

This voluminous record of successful operating experience with zirconium has been and continues to be supported by an extensive in- and ex-reactor test program in which the limits of applicability have been explored and continuously refined. In contradiction therefore to Gulbransen's statements, this experience demonstrates that the "inherent material problems associated with zirconium alloys" (whatever they may be) have in fact been successfully overcome in current reactor designs.

N. J. Palladino

State College, Pa.

Correction

In "Nuclear Reactor Safety: Further Points of Clarification" by Frank von Hippel in the January 1976 issue of the *Bulletin*, a phrase was inadvertently omitted. In the discussion of *The Linear Hypothesis and the Consequences of Low Doses of Radiation* on page 45, the sentence should have read: "This makes it possible to extrapolate from the observed effects of radiation at high doses and dose rates *to lower doses and dose rates* such as those which would occur far downwind from a reactor accident."

DOOMSDAY ALTERNATIVE

Everyone has cause for alarm reading articles in many magazines about runaway spread of nuclear weapons, pervasive deterioration of the human habitat, and other global crises. But there is a way out: The creation of a democratic world federation which could provide rational means for solving global problems, not possible while militarized nations remain sovereign.

But—how? Welcome news is provided by an international group working on an up-dated constitution for world government supervised by an elected parliament. The first draft is circulating for comments, titled: **A Constitution for the Federation of Earth.**

Three thousand $10 contributions are needed now to reconvene the drafting commission (half from developing countries) for intensive work on final draft, to be submitted to a World Constituent Assembly called for June 1977 at Innsbruck, Austria. More $ are needed for the Constituent Assembly. After adoption at the Assembly, the world constitution will be submitted to the people and nations of Earth for ratification.

Obtain details including Call to Constituent Assembly from: **World Constitution & Parliament Association,** 1480 Hoyt St., Lakewood, Co. 80215. $10 contributors will receive copy of preliminary draft.

Applications Invited for AAAS Congressional Science Fellowships

The American Association for the Advancement of Science invites applications for the fourth consecutive year of its Congressional Science and Engineering Fellow Program.

Detailed information on the application procedure and other information about the program are available from Dr. Richard A. Scribner, Director, AAAS Congressional Science Fellow Program, AAAS, 1776 Massachusetts Avenue, NW, Washington, D.C. 20036. The deadline for application is 31 March 1976.