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UNITED STATES
NUCLEAR REGULATORY COMMISSION
WASHINGTON, D.C. 20555-0001

August 7, 1995

Mr. Lake Barrett, Deputy Director
Office of Civilian Radioactive
Waste Management
U.S. Department of Energy, RW-2
1000 Independence Avenue, SW
Washington, DC 20585

SUBJECT: REVIEW OF POTENTIAL FOR UNDERGROUND AUTOCATALYTIC CRITICALITY

Dear Mr. Barrett:

In accordance with my letter to you of May 1, 1995, the U.S. Nuclear Regulatory Commission has completed a review of reports written by scientists at Los Alamos National Laboratory (LANL), as well as related reports by scientists at the Savannah River Laboratory, Lawrence Livermore National Laboratory (LLNL), and other sources, which discuss the potential for criticality of fissile materials in deep geologic disposal. Our review focused primarily on the paper written by Drs. C. D. Bowman and F. Venneri of LANL, entitled "Underground Supercriticality from Plutonium and Other Fissile Material." The Bowman paper postulates that a supercritical reaction could occur in a repository that would release sufficient energy that it would constitute a "nuclear explosion." As further explained in this letter, we do not believe a supercriticality event as postulated in the Bowman paper is credible, nevertheless criticality prevention is a public health and safety issue that DOE needs to address in its license application.

NRC has always considered controls to prevent accidental criticality to be an important issue with respect to the disposal of high-level radioactive waste in a geologic repository. As you are aware, NRC regulations already include specific requirements designed to preclude any accidental criticality, much less a supercritical reaction. It was within this context that the staff reviewed the Bowman paper. A review team led by Dr. Michael J. Bell, Chief of the Engineering and Geosciences Branch of the Division of Waste Management, has developed the enclosed set of comments which I am forwarding to you for U.S. Department of Energy (DOE) consideration.

First and foremost, NRC staff does not consider that the scenarios and conditions postulated in the Bowman paper are shown to be credible for any realistic repository situation. Staff's technical basis for this position on the Bowman paper is presented in the enclosure. Specifically, NRC staff found that the Bowman paper assumes some idealized conditions which cannot exist in a repository and which must be modified to be credible. For example, the assumptions of homogeneous mixtures of pure water, silicon dioxide and plutonium-239 in spherical geometry are not credible in an actual repository. Furthermore, other conditions assumed by the Bowman paper, while possible, have very low probabilities of occurring that need to be considered. Examples are the probabilities that sufficient water will contact the waste packages to expose the waste form to leaching and that neutron absorbing material in the

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waste packages will be selectively transported out of the system. This analysis will be very site specific and will differ greatly for an arid site like Yucca Mountain from the generic calculations of the Bowman paper.

Notwithstanding the above, DOE, as the future applicant, has the burden to demonstrate that what they propose is safe and meets NRC requirements. DOE must provide, in its license application, rigorous, site-specific technical analyses of repository criticality safety. Such analyses, assessing ranges of credible scenarios and mixtures of fissile and other materials, would place additional perspective on the merits of the Bowman paper and its relationship to DOE's activities at Yucca Mountain, Nevada. DOE scientists have particular expertise in designing and analyzing supercritical systems that can be applied to determine whether, under realistic repository conditions, autocatalytic criticality would ever be possible. An integrated systems and multi-disciplinary approach to criticality analyses is suggested, with consideration of the relationship of the assumptions and results to criticality control design decisions. NRC recognizes that DOE's Office of Fissile Materials Disposition (OFMD) has yet to decide on a method of disposition of weapons related materials, and that much of the analysis in the Bowman paper could, therefore, be considered premature.

The observations, presented in the enclosure, present the review team's technical basis and logic for determining that the conclusions of the paper by Drs. Bowman and Venneri are not, as currently developed, expected to present a significant obstacle to repository licensing. The review team results have been divided into two sections, corresponding to those considerations relevant to the current repository program, in Part I, and those items not viewed as relevant at this time, in Part II. The relevance of a particular consideration was determined based upon factors, such as its applicability to wastes already being considered for disposal (e.g., high-enriched spent fuel), or its potential to occur within the regulatory time frame of interest. If a decision is made to dispose of excess weapons plutonium or surplus highly enriched uranium in an NRC licensed repository, or if the regulatory time frame of interest is extended, some of the items in Part II would need to be considered. However, such consideration should reflect the limitations on criticality imposed by meeting NRC regulations.

Though NRC has yet to formally review DOE efforts to address criticality controls for a repository, based upon technical interactions between NRC and DOE staff, and the literature to date documenting DOE efforts, NRC considers that all of the considerations listed in Part I of the enclosure are, to some extent, being addressed in DOE's repository program. Therefore, the review team has offered the enclosed considerations and suggestions primarily to provide additional clarification of NRC expectations for criticality analyses, which DOE may find to be useful for future work in this area. We also suggest that the Office of Civilian Radioactive Waste Management share the review team observations with other DOE offices who may be working in this area.

The NRC review team found the LLNL paper compiled by Dr. R. A. Van Konynenburg, which questions the conclusions of Drs. Bowman and Venneri, to be a fairly comprehensive, though somewhat qualitative, treatment of the

L. Barrett

autocatalytic criticality issue. DOE may wish to use this report as a basis from which to perform more detailed quantitative analyses, as described above.

As I suggested in the letter of May 1, 1995, DOE may want to consider broadening the scope of DOE's planned "Report on Criticality Control for Disposal of Spent Nuclear Fuel" to address additional criticality control considerations, such as those discussed herein and in the aforementioned DOE laboratory reports.

My staff and I stand ready to discuss criticality matters with you at any time. If there are any questions related to this subject, please contact me on (301) 415-7800 or Dr. Michael Bell on (301) 415-7286.

Sincerely,

Carl J. Paperiello, Director
Office of Nuclear Material Safety
and Safeguards

Enclosure: As stated

Paper proofed by C.Jensen 7/12/95

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NRC Staff Comments on the Report
"Underground Supercriticality From Plutonium and Other Fissile Material"
by Drs. C. D. Bowman and F. Venneri
Los Alamos National Laboratory

Introduction

Drs. C. D. Bowman and F. Venneri, two scientists from Los Alamos National Laboratory (LANL), have published a technical paper (herein referred to as "the paper") entitled "Underground Supercriticality from Plutonium and Other Fissile Material,"[1] which theorizes that a sequence of events and conditions are possible which could lead to significant energy release from criticality excursions by fissile materials in geologic disposal sites. Although the paper primarily applies to the proposed disposal of weapons grade plutonium and highly enriched uranium, the authors qualitatively state that the arguments can be extended to all fissile materials buried in a repository.

The U.S. Nuclear Regulatory Commission established a review team to assess the technical content of the paper and other reports on the subject. The review team included NRC staff and contractor personnel from the Center for Nuclear Waste Regulatory Analyses (CNWRA). The objectives of the review team were to: (1) conduct a broad-based, multi-disciplinary review of the potential for and consequences of autocatalytic reactions in a geologic repository; (2) focus the review on credible scenarios and mechanistic processes and events; (3) conduct scoping analyses to verify the paper's calculations and conclusions and to assess the various options available to the Department of Energy's (DOE's) Office of Fissile Materials Disposition (e.g., co-vitrification/dilution of weapons materials with high-level waste); and (4) develop comments, questions, and issues to forward to DOE to ensure that technical issues raised by the paper, which should be considered in licensing of a geologic repository, are addressed in a timely and appropriately sound scientific manner. As NRC's primary concern is with those facilities that it will license, the review team focused its efforts on the proposed geologic repository site at Yucca Mountain, Nevada.

The NRC review team does not consider that the scenarios and conditions postulated in the Bowman paper are shown to be credible for any realistic repository situation. Specifically, the review team found that the Bowman paper assumes idealized conditions which cannot exist in a repository and which must be modified to be credible. Furthermore, other conditions assumed by the Bowman paper, while possible, have very low probabilities of occurring that need to be considered. Notwithstanding the above, DOE, as the future applicant, has the burden to demonstrate that what they propose is safe and meets NRC requirements. DOE must provide, in its license application, rigorous, site-specific technical analyses of repository criticality safety. Such analyses, assessing ranges of credible scenarios and mixtures of fissile and other materials, would place additional perspective on the merits of the Bowman paper and its relationship to DOE's activities at Yucca Mountain, Nevada. DOE scientists have particular expertise in designing and analyzing supercritical systems that can be applied to determine whether, under realistic repository conditions, autocatalytic

Enclosure

criticality would ever be possible. Specific technical observations identified by the review team during the review of the paper and its related literature, have been provided below. These items discuss observations of the review team, and present the review team's technical basis and logic for determining that the conclusions of the paper by Drs. Bowman and Venneri are not, as currently developed, expected to present a significant obstacle to repository licensing.

The review team found the Van Konynenburg paper[2] written at Lawrence Livermore National Laboratory (LLNL), which questions the conclusions of Drs. Bowman and Venneri, to be a fairly comprehensive treatment of the autocatalytic criticality issue. However, the review team noted and would like to emphasize the qualitative nature of several arguments made in the Van Konynenburg paper (and of several other papers which criticize the findings of Drs. Bowman and Venneri). DOE may wish to use the Van Konynenburg paper as a basis from which to perform more detailed, quantitative analyses in future work(s) related to this issue. NRC suggests that a systematic approach be adopted to help ensure the adequacy of any future efforts to analyze the progression of postulated underground criticality events.

NRC recognizes that DOE's Office of Fissile Materials Disposition (OFMD) has yet to decide on a method of disposition of weapons-related materials, and that much of the analyses of the paper could therefore be considered premature. However, in Part I, below, the review team has listed several considerations, relevant to the current repository program, which should be considered by DOE in continuing development of its repository program. Based upon technical interactions with DOE and the literature to date documenting DOE efforts to characterize repository criticality, staff considers that all of the considerations listed in Part I are, to some extent, being incorporated into the DOE repository program. These considerations and suggestions are offered primarily to provide additional clarification of NRC expectations for criticality analyses. The relevance of a particular consideration was determined based upon factors, such as its applicability to wastes already being considered for disposal (e.g., high-enriched spent fuel), or its potential to occur within the regulatory timeframe of interest.

Additional items are presented in Part II, below, which are not viewed as relevant to the current repository program at this time. We suggest that the Office of Civilian Radioactive Waste Management (OCRWM) share these, as appropriate, with other DOE offices who may be working in this area. If a decision is made to dispose of excess weapons plutonium, or highly enriched uranium, in an NRC licensed repository, or if the regulatory timeframe of interest is extended, some of these items may also need to be considered. However, such consideration should reflect the limitations on criticality imposed by meeting NRC regulations.

Finally, it should be noted that, as previously stated, this review effort focused primarily on the paper by Drs. Bowman and Venneri, and its related literature, and the review should not be construed as an effort to identify all criticality issues related to the licensing of a deep, geologic repository for disposal of high-level waste.

Part I. Considerations Relevant to the Current Repository Program

In this Part, the review team has listed several considerations, relevant to the current repository program, which should be considered by DOE in continuing development of its repository program. Based upon technical interactions with DOE and the literature to date documenting DOE efforts to characterize repository criticality, staff considers that all of the considerations listed in Part I have, to some extent, been incorporated into the repository program. These considerations and suggestions are offered primarily to provide additional clarification of NRC expectations for criticality analyses. The relevance of a particular consideration was determined based upon factors, such as its applicability to wastes already being considered for disposal (e.g., high-enriched spent fuel), or its potential to occur within the regulatory timeframe of interest.

1.1. *General recommended approach.* Analyses assessing ranges of credible scenarios and mixtures of fissile and other materials should be completed before the physical possibility of energetic autocatalytic nuclear excursions for fissile materials in a repository is accepted or dismissed. Probabilities of initiating events for situations resulting in sudden large reactivity insertions should be incorporated into analyses. This approach should be performed with consideration of the relationship of assumptions and results to criticality control design decisions. A systematic and multi-disciplinary approach is recommended. In order to provide information useful for decision-making, physical modeling of situations related to waste criticality should be confined to selected situations, each of which address a physically realistic scenario from beginning to end. The logical organization and delineation of the scenarios analyzed should be based on risk and decision theories.

Recommended transient modelling approach. To adequately analyze the neutronics and energetics of hypothetical underground criticality events, it is essential to identify and model the important phenomena and processes that could affect the outcome. If a need is found to characterize the potential for explosive excursions in a given representative critical configuration, static modelling and neutron transport calculations (using codes such as KENO-Va, MCNP, ANISN, or ONEDANT) should be performed to quantify reactivity feedback coefficients and other kinetic parameters (e.g., neutron lifetime) as functions of material densities and temperatures of interest. The resulting information should be processed and used in a coupled simulation of neutron kinetics and thermal hydraulics/hydrodynamics using representative moderator and reflector conditions. (Although the modification of a weapons code for this purpose has been suggested, NRC recognizes concerns for such an approach due to classification considerations and the associated constraints on open discussion and peer review. Alternatively, it may prove easier to develop an entirely new code.)

1.2. *Extension of arguments to non-weapons materials.* The paper qualitatively argues that its conclusions can be extended to spent nuclear fuel (SNF) from high enriched uranium (HEU) fueled research reactors and commercial reactor SNF. Also mentioned

are navy fuel and mixed oxide fuel resulting from the use of weapons plutonium in commercial reactors (another option being considered by DOE's Office of Fissile Materials Disposition (OFMD)). This extension is attributed to the higher solubility of uranium with respect to plutonium and the decay of the Pu-240, both of which will contribute to making the spent-fuel isotopic composition "approximate the isotopic composition of w-Pu and the criticality risk from stored material will become larger as more time passes."[1] The increasing reactivity of intact SNF in time, which exhibits a local maximum near 20,000 years, due to the decay of Pu-240, has been documented.[3] Staff suggests that DOE incorporate into disposal criticality analyses, consideration of the criticality potential in time for spent fuel as a function of its changing composition due to possible preferential leaching of actinides or fission products. Staff further suggests that such efforts focus not only on the waste form, but also on the criticality potential of actinides leaving the waste form, which, for example, could subsequently experience some type of reconcentration mechanism.

1.3. *Situations resulting in gradual reactivity insertions.* A more rigorous neutronic analysis, than done in the paper, is suggested, which would allow for the adequate characterization of competing feedback effects over the range of material mixtures and scenarios of interest. Such an analysis has not been provided and should be completed before one accepts or dismisses the physical possibility of energetic autocatalytic nuclear excursions arising from gradual, migration-induced insertions of reactivity.

References 2 and 4 have pointed out that the nuclear excursion analyses performed by Bowman and Venneri failed to account for the potentially stabilizing effects of thermal expansion. As part of LANL's internal review process,[4] Dr. G. H. Canavan performed a simplified analysis[5] asserting that negative neutronic feedback from thermal expansion would always prevent or override the development of positive feedback mechanisms in a gradual approach to criticality. Specifically, it was claimed that thermal expansion, arising from small temperature increases of a few degrees or less, would terminate such criticality events within seconds or minutes. However, Dr. Canavan's analysis, which was based on criticality being produced by the outward diffusion of cold fissile material in a dry-rock medium, contained several questionable assumptions, key among which was the assumed negative value of the temperature feedback coefficient. While it is true that most well-moderated critical systems tend to have negative temperature coefficients, there are many notable exceptions. Furthermore, although not addressed in that analysis, one could also infer that recriticality could occur upon cooling down and that the recriticality temperature would gradually increase with each heating and cooling cycle as materials continue to migrate. It is well known that many critical systems tend to develop positive temperature feedback components at higher temperatures. Such positive feedbacks could arise from boiling of water in wet mixtures, neutron-spectral heating into the Pu-239 fission resonance in wet or dry mixtures, or any of several other autocatalysis mechanisms discussed in References 2 and 6.

The paper ignores, or mentions, but neglects, some important phenomena affecting the neutronics and nuclear physics of the assumed and analyzed systems. NRC staff offers the following considerations which could be incorporated into analyses to properly characterize the reactor kinetics parameters for the described systems:

(a) Secondary constituents (e.g., trace elements such as sodium) in the rock and water may reduce reactivity and affect feedback (see Item 2.2, below);
(b) Thermal expansion of water relative to rock at and below saturation levels in rock can provide dynamic feedback (identified by the paper as a mechanism for positive feedback in wet overmoderated scenarios). This effect is less important for unsaturated rock;[a]
(c) Low-energy resonance of Pu-239 (0.35 eV), not modelled in the paper, is an important quasi-prompt contributor to positive feedback at moderate-hot temperatures and is a negative feedback contributor at extreme (bomb-like) temperatures;
(d) Low-energy resonance of Pu-240 (1 eV), not modelled in the paper, reduces cold k_{eff} until it sufficiently decays (i.e., until about 20,000 years);[3]
(e) Decay of fissile materials (e.g., Pu-239 to U-235) over extended time frames may reduce reactivity; however, Parks, Williamson, and Hyder[7] have done calculations for Pu-239/Pu-240/U-235/U-236 mixtures which show that the infinite multiplication for a system does not significantly change (<0.1 change in K_{inf}) over at least 100,000 years;
(f) Doppler broadening and self-shielding in fuel and moderator resonances due to heating should not be neglected, because this phenomenon may give important feedbacks at "hot" temperatures, especially for larger fuel particles and less dilute mixtures. Further, this effect may be more pronounced for water moderated systems;
(g) Neutron migration area and leakage, as a function of material densities and temperatures, is mentioned in the paper and analyzed with simplifying assumptions (e.g., 1/v dependence of cross sections). A rigorous calculation may be needed to properly quantify this potentially important feedback mechanism;
(h) Fissile depletion and fission product ingrowth should not be discounted for slow approaches to criticality (such as those induced by geochemical effects);
(i) Heating or hardening of the neutron spectrum can produce positive feedback in Pu-239 at moderate temperatures and negative feedback from Pu-240 at great temperatures;
(j) Radiolytic-vapor expansion of water, and eventually of molten materials, can provide positive or negative prompt feedback; and
(k) The homegeneity assumption of the fuel/moderator mixture in the paper could overestimate calculated k_{eff}.[8]

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a. This refers to the fact that thermal expansion of liquid water in a saturated or supersaturated mixture will expel or extrude water away from the local matrix, resulting in a modified moderator mixture. This is not true for an unsaturated mixture, where water has room to expand within the material matrix.

1.4. *Characterization of reactivity feedback parameters.* The dry autocatalytic condition is a prerequisite to the thesis of significant energy releases (explosions) occurring and affecting nearby containers. For conditions in which water is present and is driven off by heat from criticality, the feedback can be estimated for the assumed conditions from Figure 1 of the paper (although this is questioned, see below). Once a dry condition is reached, Figure 1 can no longer be used to characterize reactivity feedback. Except for the "emplacement in the ear region" scenario discussed below in Item 2.2., the arguments in the paper showing that a critical system can have a positive feedback coefficient in a dry state are unclear to the staff. Important elements of reactivity feedback, such as those mentioned in Consideration 1.3, are ignored, or mentioned but neglected, by the paper for progressing transients. An initially positive (or negative) feedback could become overwhelmed by emergent temperature-dependent spectral and density related feedback mechanisms (positive or negative). As insufficient information has been presented to properly characterize transient behavior in super-critical dry rock/fissile material systems, NRC staff believes that further justification of the paper's thesis through additional analyses and calculations is necessary.

Autocatalysis effects for realistic rock. Calculations done in support of performance assessment for disposal of HEU fuel in tuff[9] have shown that in a highly saturated environment, a spherical assembly in realistic rock which becomes critical and generates sufficient heat will shut itself down as water is removed (i.e., shows negative feedback). This self-shutdown mechanism was deemed too large to allow a criticality resulting in a large fission release (between 10^{18} and 10^{21} fissions). Preliminary calculations done by the CNWRA after the 200 cm sphere case of Figure 1 in Drs. Bowman and Venneri's paper, show that when a sphere of realistic rock bearing plutonium "dries out," the reactivity of the sphere decreases (directly contradicting Figure 1 of the paper). Staff suggests that more work is needed to better assess the dynamics of how realistic rock systems react when drying, and thus how representative of rock a pure SiO_2 system is. The paper's simplistic analytical treatment of the material properties of moderator and reflector materials and system geometry is viewed by the staff to be potentially grossly over-conservative and can introduce possible conceptual errors.

1.5. *Characterization of mass transport mechanisms.* The criticality scenario requires Pu release from waste forms, transport, and deposition in a rock enclosure. None of these processes are described in detail by the paper. Table A1 of the paper provides solubility values for relevant constituents in water at 300 K; however, no consideration of the dependence of solubility on system chemistry has been incorporated. Wilson, et. al.,[10] cited as a primary reference in the paper, clearly indicates that solubility is a function of system chemistry, and gives expert opinion ranges of solubilities ranging from 10^{-10} to 10^{-6} molar for Pu, and from 10^{-8} to 10^{-2} molar for U (not UO_2 as indicated in the paper). Although differences in solubilities are important factors for relative release calculations, glass dissolution does not occur by independent release of separate compounds according to the relative solubilities of pure phases. Releases will be interdependent and dependent upon system chemistry.

Preliminary calculations by the CNWRA using EQ-3[11] with conditions loosely representative of Yucca Mountain yielded Pu solubility of 10^{-12} molar. At a solubility of 10^{-6} molar, 63,000 m^3 of plutonium-saturated water would have to pass through a 100 cm radius sphere to transport 15 kg of Pu by means of dissolution and re-deposition. Scenarios or mechanisms which could generate this amount of water contact and subsequent deposition seem unrealistic and have not yet been identified.

Staff recommends that detailed simulations of releases for representative geochemical conditions be modelled, using an established code such as EQ-3/6,[12] to estimate relative and cumulative releases of the various compounds within the waste form(s). Staff further recommends that DOE's criticality studies incorporate the possibility and effects of colloidal transport of Pu coupled with physical filtration at a point of changing hydraulic properties in rock as a mechanism for transport and reconcentration.

1.6. *Identification of alternative redistribution mechanisms.* Realistic scenarios for creating autocatalytic configurations are not discussed in the paper. Staff believes that redistribution methods other than transport in dissolved ionic species should be identified and assessed (see the colloidal transport discussions in Considerations 1.5, above, and 1.7, below). Redistribution by waste form collapse, as mentioned in the paper as a possible means of large reactivity insertion, should be quantitatively assessed (both in terms of likelihood and reactivity worth). Transport by slurry would seem highly unlikely in a partially saturated environment barring a large fracture intersecting a waste package which allows infiltration to quickly penetrate and pass over the waste form. This mechanism may require complete degradation of the package to occur.

Certain alternative distribution mechanisms could be theorized to result in sudden, potentially large, nuclear reactivity insertions. In fact, the sudden mechanical insertion of reactivity implicitly entails an extra event (such as rock collapse, water ingress, etc...) in or near fissile materials that may or may not otherwise follow a gradual approach to criticality. Depending on the details of such an event and configuration, such a relatively sudden event could not only add or remove reactivity, but could also enhance or suppress any autocatalytic nature of the subsequent neutronic feedback effects. In estimating the probability of sudden mechanical reactivity additions, it is necessary to consider the credibility and frequency of initializing events (e.g., earthquakes, hydrologic transients, stressing of rock due to heating/steam, etc...). Reference 13 cites evidence that the Oklo natural analog reactors were ultimately shut down by collapse of rock and displacement of water (a large negative reactivity insertion, in that case). Without due consideration of the probabilities of such initiating events, it seems clear that a sudden positive reactivity insertion to certain underground mixtures of fissile material could indeed result in a prompt supercritical, initially autocatalytic excursion. However, it is expected that dynamic analyses will show that consequences of such transients are less violent than projected in the paper.

1.7. *Potential geochemical effects of iron.* The repository will contain a significant amount of iron in the form of steel structural supports, waste packages, and perhaps backfill.[14] The corrosion of these materials will generate iron oxyhydroxide phases, which often exist in colloidal form. These colloids may serve as substrates for sorption of dissolved and colloidal forms of Pu.[15] Transport of colloids from the underground facility has not yet been shown to be an insignificant mechanism affecting release of radionuclides to the geologic setting. If transport of Pu by colloids proves to be significant, dispersal, rather than concentration of Pu, should be expected.

Iron has been shown to increase the rate of dissolution of borosilicate glass.[16] The effect of an increased rate of dissolution on the transport and concentration of Pu will be dependent upon the flow rate of fluids and rate of precipitation of secondary phases. Little work has been done to date to assess the coupled effects of dissolution and transport. However, this coupling is a key component of any scenario in which Pu is mobilized from a waste form.

Iron has also been shown to reduce the k_{eff} of a plutonium (or uranium) and SiO_2-H_2O system by thermal neutron absorption.[17] In criticality analyses of low-level waste disposal sites, the presence of 12-gauge or carbon-steel containers reduced the reactivity of the Pu (or U) and SiO_2-H_2O system. Reactivity was further reduced when water was introduced to the configuration. Staff recommends that DOE consider the effects of iron on the reactivity of the Pu-U system. DOE should also consider effects of iron on the solubility, transport, and dispersal of Pu and U.

1.8. *Phase transitions in host rock.* Appendix B of the paper indicates that "if the pressure reaches 30 GPa [300 kbar] the rock completes a phase change to stiskovite (sic)."[1] At elevated pressures silica undergoes a phase change to stishovite which has been observed at meteorite impact sites, and synthesized in laboratories at high temperatures and pressures (13 GPa and > 1200 °C).[18] Although the rock at Yucca Mountain is about 70 percent silica by mass, pure silica phases occur in percentages of 0 to 15 or 20 percent. Many other phase transitions would occur in a natural rock before stishovite forms from a fraction of the phase assemblage. The transition from another silica phase to stishovite may occur at 300 kbar at high temperatures because it is temperature dependent. However, transition as treated in the paper is temperature independent. The pressure-density diagram, Figure B1 in the paper, is asserted to be a phase diagram for Westerly granite (though unreferenced). As used, an unrealistic temperature independent phase change has been imposed on the rock with respect to stishovite formation. This transition is asserted to have a major impact on explosive yield. Staff suggests DOE incorporate realistic phase transitions for a multiphase assemblage in estimates of rock deformation and/or explosive yield.

Changes in temperature (and the rates of changes in temperature) will affect the thermal and mechanical properties of the rock/water/fuel system (as well as the neutron spectrum of the system and the absorption properties of the materials). Among other things,

compressive strength is a function of temperature and saturation, and the thermal conductivity of saturated or wet tuff is significantly higher than that of dry or dehydrated tuff.[19] The variation of properties as temperature changes and water "burns off" should be assessed. Rock subjected to such heating would be expected to exhibit some sort of elastic or inelastic deformation that would have a dynamic effect on the reacting system. Such deformation may potentially control oscillatory behavior in gradual onset criticality scenarios. Also, the heterogeneity of the rock may provide for pathways of least resistance through which such high temperature and pressure gradients would be expected to cause movement of reacting system constituents.

Part II. Review Team Technical Observations, Which Are Not Relevant to the Current Repository Program at This Time.

The following items discuss observations of the review team, and further present the review team's technical basis and logic for determining that the conclusions of the paper by Drs. Bowman and Venneri are not, as currently developed, expected to present a significant obstacle to repository licensing. However, these items are not viewed as relevant to the repository program (e.g., they may apply only to disposal of weapons materials). We suggest that OCRWM share these observations, as appropriate, with other DOE offices who may be working in this area. If a decision is made to dispose of excess weapons plutonium or highly enriched uranium in an NRC licensed repository, or if the regulatory time frame of interest is extended, some of these items may also need to be considered in OCRWM's development of the repository.

2.1. *Time frame considerations.* The authors concede that large amounts of water and great lengths of time are required to establish the conditions analyzed in the paper. Time, in series and parallel, is required to compromise container integrity, decay poisons (25,000 years is mentioned), and selectively leach, transport and deposit fissile, and other materials. Though the authors could argue that such processes are inevitable, the quantification of relative consequences from such events should be made in light of the overall performance of the facility. Staff believes that assuming a particular end state for analysis without consideration of factors contributing and leading to that end state to be of little utility in assessing overall facility behavior and potential impacts to public health. By employing a systematic, multi-disciplinary approach, the staff believes it may be possible to demonstrate that successful performance (i.e., in terms of the performance objectives or designed criticality control) will continue for at least 10,000 years, and perhaps indefinitely. Such analyses have yet to be done in the repository program.

Natural and experimental analogs exist which can provide valuable, if limited, insight into the conditions leading to, and consequences of, various types of criticality excursions. The Oklo reactors of Gabon may be useful as a natural analog of slow, geochemical processes leading to criticality. For faster reactivity insertions, information may be gleaned from criticality accident case histories and solution criticality experiments, such

as the CRAC series of experiments and others.[20] Additional discussion of natural and experimental analogs is provided in Reference 9.

2.2. *Pure water and SiO_2 as moderating and reflecting materials.* Rock in the proposed repository at Yucca Mountain, Nevada, is expected to contain significant quantities of elements such as potassium and sodium which have better neutron absorption properties than silicon, and trace quantities (less than 1 wt. percent) of even greater absorbers, such as titanium and chlorine.[21] Dissolved materials in groundwater also may have absorptive properties. Because these materials impact the neutron economics and dynamic response of a system, the staff believes it is appropriate to include them in analyses. The thermal expansion of the rock matrix is expected to be a significant delayed feedback mechanism, especially for quasi-stable scenarios involving slow approaches to criticality, such as those due to geochemical phenomena. Analyses taking credit for such processes will be quite complicated due to factors such as constraint of expansion by induced stresses in surrounding rock.

Near the end of the report, Drs. Bowman and Venneri estimate that 50 percent larger quantities of fissile material would be needed for "real" rock as compared to pure SiO_2. This estimate has been verified by CNWRA staff during the review. Specifically, it was found that about 116 kg Pu-239 is the critical mass when the rock compositions found in NUREG/CR-6288,[22] Table 4-1, Column 3 were used, as opposed to 70 kg for the pure Pu-239/SiO_2 system of the paper's Case C in Figure 1.

To assess the effect of trace nuclei on the system, the following scoping calculation was performed by NRC staff. Using Figures 3 and 4 from the paper by Drs. Bowman and Venneri, the peak criticality (optimum moderation) point for 50 kg Pu-239 distributed in a sphere of pure SiO_2 of density 2.2 g/cc is observed to have a k_{eff} of 1.067 at 140 cm radius. This value was obtained by NRC staff using SCALE 4.1[23] with 44-group cross sections, and agrees with the Figure in the report (in fact, the entire Figure 4 for 50 kg Pu was reproduced and showed agreement). NUREG/CR-2937[24] gives a chlorine content for Vitric Paintbrush Tuff of 0.19 weight percent (7.2E-05 atoms/barn-cm, including a 10 percent porosity). Re-calculation including chlorine in the moderator and keeping everything else constant lowers dry k_{eff} from its 1.067 peak to 0.967. Inclusion of other nuclei in tuff[24] further lowers dry (but hydrated to 3.88 wt percent water) k_{eff} to 0.712. It was not determined if these lower values represent the peak k_{eff} for the altered system. Parsons, et. al.,[8] have investigated this phenomena. Although this chlorine is in the rock, chlorine or other materials dissolved in water might have a similar effect in moist systems. However, somewhat lower Cl^- concentrations around 10 mg/L (1.7E-07 atoms/barn-cm) have been measured in the groundwater around Yucca Mountain.[24]

Formation of dry, autocatalytic condition. It should be noted that the scenario discussed in the paper describing emplacement in the "ear" region of the paper's Figure 3, and subsequent dispersion-induced dry autocatalytic super-critical conditions, could not occur in a dry medium if trace nuclei such as chlorine are introduced as done in the above

calculation. Figure 3 is the only place that the paper describes conditions leading to dry autocatalytic behavior. In the "ear" region of the Figure, as one moves vertically down the right axis, dispersion in dry rock can insert reactivity and result in criticality. NRC staff believes the region of interest of this Figure for the dry autocatalytic condition is insufficiently developed with respect to actual rock conditions. Specifically, properties of realistic rock including increased poison or hydrated water may preclude the "emplacement in the ear region" scenario, or eliminate the ear region from the Figure altogether. Parsons, et. al.,[8] have investigated this phenomena and found that the "bottom of the ear" drops out for 25,000 year-old weapons Pu in realistic rock.

2.3. *Heterogeneity of rock and engineered features.* Although the homogeneous medium assumed in the paper simplifies the analysis, its applicability on the scale of several meters is questionable for the repository emplacement area (a good discussion of this is presented by Van Konynenburg).[2] Heterogeneous mineral phases in the host rock, fractures and fissures, and the presence of engineered features such as other containers, backfill, or the drift itself, all work against the homogeneity assumption as system radius increases. These heterogenous features would make it increasingly more difficult for a system to form a spherical shape as the radius increases. It is anticipated, that if quantified, these heterogeneous, non-spherical effects will increase neutron leakage and decrease reactivity. Reference 8 indicates that, for heterogeneously distributed fuel particles, k_{eff} decreases, as compared to homogenized idealizations.

Rock porosity. A maximum water content in the systems analyzed should be established using the porosity of the host rock. The abscissa in Figure 1 in the report displays moisture content in terms of molar fraction, ranging from pure liquid ($X=0$) to a pure solid ($X=1$). Correspondingly, the porosity of the rock is not constant but varies from 100 to 0 percent. For the Topopah Spring unit in the proposed repository site at Yucca Mountain, the average porosity is 10 percent.[25] This value would limit the molar fraction of rock to values greater than 0.86 according to the paper's Figure 1. Consequently, this would limit the autocatalytic behavior described to the larger volume Pu-239/SiO_2/H_2O systems. The 10 percent porosity mentioned above is taken to be an average value, Reference 2 discusses a practical upper limit for host rock porosity of 20 percent which yields a mole fraction of water of 0.24.

The 30 cm radius critical volume of Pu-239 as shown at Point F in the paper's Figure 1 contains less than 2 kg Pu-239 and corresponds to a porosity of approximately 55 percent. Such porosity values could possibly occur in backfill materials if used, but are extremely unlikely in any repository host rock. It is expected that backfill, if used, will be designed to keep water away from waste. The impacts of as-built or degraded backfill on repository performance have yet to be assessed.

Matrix-bound water. Competing (in terms of neutron economy) with the increased absorption (and decreased reactivity) of realistic rock as compared to SiO_2 is the fact the rock may contain loosely- and tightly-bound matrix water or other hydrogen bearing

materials which could increase moderation and thus reactivity. NUREG/CR-2937[24] (after White, et. al.[21]) gives a matrix-bound water content for Vitric Paintbrush Tuff of 3.83 wt percent (13.4 mole percent). Although dehydration of the rock could occur subsequent to closure of the repository due to heating associated with fission product decay, after substantial contact with water during the geologic time frames necessary to separate plutonium, rehydration of the host rock may occur (likely to a different mineral form). Supporting this presumption is that significant water would implicitly be necessary to initiate the amount of leaching necessary to the paper's thesis, including that needed to degrade both the containers and waste forms. It should be noted that altered mineralogy would also impact properties such as porosity, permeability, and compressive strength.

Analyses which start from a completely dry state as described in "Case A" for Bowman-Venneri Figures 1 and 3 would seem unrealistic because significant water ingress (causing rehydration) would occur prior to substantial leaching of the waste forms.

2.4. *Criticality of U-235 progeny.* Pu-239 decays to U-235 with a half life of 24,100 years. The fissile U-235 is likely to exhibit considerably different behavior in the environment than plutonium. Uranium is generally much more soluble and sensitive to the oxidation/reduction potential of its environment. One can postulate mechanisms for transport, reconcentration and criticality of U-235 that would be much less likely for Pu-239. Upon failure of the waste package, the glass waste form containing the weapons Pu-239 will begin to dissolve at a rate determined by the degradation rate of the glass and the flow rate of liquid water in contact with it. Another potentially large source of U-235 is highly enriched uranium fuel, such as that from the Advanced Test Reactor. U-235 will dissolve from the waste form (or from previously released plutonium) at a rate determined by the solubility of secondary minerals and the flow rate. Migration of the uranium could proceed with the flowing groundwater until it encounters a zone that is sufficiently reducing to cause precipitation. This scenario would be similar to actual processes in nature responsible for the formation of uranium ore deposits, i.e., "roll fronts."

Such a scenario would be similar in some ways to the Oklo natural fission reactor, in which uranium enriched to 2-3 percent U-235 concentrated in a roll front deposit. However, the uranium resulting from the decay of weapons-grade plutonium would be nearly 100 percent U-235. In addition, the uranium deposit in the present scenario could result from the merging of the releases from a number of waste logs. The uranium situation is fundamentally different from Pu-239 criticality portrayed in the paper by Drs. Bowman and Venneri, because staff believes that mechanisms in the latter which concentrate the plutonium in a single place are insufficiently developed.

While Yucca Mountain is considered to be an oxidizing environment, the potential for the U-235 reconcentration scenario should, nonetheless, be analyzed. Among the factors that should be considered in analyzing the potential for and consequences of this alternative criticality scenario are: (1) the effects of U-235 and U-238 released from spent-fuel

canisters; (2) the stability of the glass waste form in the silica-rich Yucca Mountain environment; (3) the relative rates of glass degradation, transport, radioactive decay, and the time frame over which the processes could be operative; and (4) hydrogeologic effects such as preferential flow through fractures in the host rock.

2.5. *Reconcentration required for small systems.* Assuming that the size of the vitrified w-Pu logs will be the same as proposed for vitrified high level waste (HLW) logs (i.e., 2 foot diameter by 10 foot length right circular cylinders), an approximately equivalent volume to the cylindrical log would be a sphere of radius 60 cm. Any sphere smaller than this radius would require reconcentration of the plutonium through a yet unidentified mechanical or geochemical process. As discussed above, staff suggests the likelihood of such processes be incorporated into scenario risk assessments.

2.6. *Potential for designed criticality controls.* Criticality control using administrative limits is possible for vitrification of weapons plutonium and also for co-vitrification of weapons plutonium with high-level waste (HLW). Co-vitrifying weapons plutonium with HLW in a yet to be built facility is one option being considered by OFMD. It seems that lower plutonium waste loadings than discussed in the paper (or large container spacings for non-co-vitrified weapons Pu logs) are technically achievable. For example, the most recent Oak Ridge Integrated Data Base Report[26] estimates that 7,990 HLW containers will be produced at Hanford by 2020. Because the volume of plutonium is negligible compared to the volume of HLW logs which will already be generated, design loadings much lower than those considered in the paper are possible for co-vitrification. For example, co-vitrifying 50 metric tonnes of weapons Pu with HLW into 5000 2'x10' cylindrical glass logs would result in around 10 kg Pu per log.

Figure 4 of the paper shows that for a pure SiO_2 moderated and reflected system, with 50 kg Pu-239, optimum moderation occurs for a sphere of radius 140 cm with a peak k_{eff} of about 1.067. The peak k_{eff} and the radius of optimum moderation decrease with decreasing plutonium content, implying benefits for lower waste loadings. Figure 4 from the Bowman-Venneri report was reproduced by NRC staff using SCALE-4.1[23] with 44-group cross sections for 50 kg Pu. The paper's and NRC's calculations agreed well. A similar curve was produced for 10 kg. The results indicated a radius of optimum moderation for 10 kg of 95 cm with a peak k_{eff} of 0.7375. As this simple calculation shows, an amount of plutonium per waste form could be defined which would remain subcritical in any of the idealized conditions analyzed in the paper (this amount is around 25 kg Pu-239 per log for the pure SiO_2 systems analyzed by the paper). A different, possibly greater, amount for more realistic conditions could be determined.

Another consideration for designed criticality control is the potential inclusion of additional, low-solubility neutron absorbent materials (i.e., beyond the boron in the "normal" borosilicate glass) into the glass matrix. Reference 2 indicates the availability of absorbers with a range of solubilities, including elements from the lanthanide series (e.g., gadolinium) and hafnium. Reference 7 suggests the use of two or more types of

absorbers (i.e., depleted uranium and gadolinium) to cover the different solubilities of uranium and plutonium, but discusses limitations related to waste loading and laboratory verification of performance. Low solubility neutron absorbent materials may also be included in designed backfill to further minimize the potential for a criticality event.

2.7. *Thermal neutron diffusion coefficient approximation.* In Appendix B of the paper, the authors have used an approximation for the definition of the thermal neutron diffusion coefficient (*D*) and hence, diffusion length (*L*) in their Equations (5) and (6). Staff believes the adequacy of this approximation for realistic conditions needs to be established.

The authors substitute a Σ_a in place of Σ_{tr} in the defining equation for *D*:

$$D=\frac{1}{3\Sigma_{tr}}$$

where

$\Sigma_{tr} = \Sigma_a + (1 - \mu_o)\Sigma_s$,

Σ_a = the macroscopic absorption cross-section of the media,

Σ_s = the macroscopic scattering cross-section of the media,

and

μ_o = the cosine of the average scattering angle.

The expression for *L* would then be:

$$L=\sqrt{\frac{1}{\Sigma_a(\Sigma_a+(1-\mu_o)\Sigma_s)}}$$

Since the situation that the paper is attempting to describe is fairly sensitive to the diffusion coefficient, the authors should show that:

$\Sigma_{tr} \approx \Sigma_a$ [i.e., that $\Sigma_a << (1 - \mu_o)\Sigma_s$ and $(1 - \mu_o) \approx 1$] for the paper's approximation to be valid.

While NRC staff believes that the demonstration of this approximation is rather straightforward for SiO_2, this is not necessarily true for realistic rocks and also for Pu-239/Pu-240 systems. The approximation breaks down for media where absorption becomes an appreciable mechanism for limiting the distance a neutron can travel into the media (e.g., the presence of a Pu absorber may alter the diffusion length).

2.8. *Energy deposition/heat transfer modelling.* In the paper, fission fragment energy is assumed to be deposited in the fissile material only. Other materials are heated only by neutron and gamma irradiation. This assumption impacts the calculations of yield for explosions and is the basis for the plutonium vaporization model. The adequacy of this assumption needs to be further addressed. The size of the fuel particles will impact the validity of this assumption; for example, in small fuel particles fission fragment heating of adjacent materials may prove significant, due to the large surface area to volume ratio which is available, and consequent increase in fission product energy deposition (and heating) in the moderator. Also, radiative and conductive heat transfer are assumed to be negligible by the paper, without quantitative justification. Reference 27 criticizes the paper's neglect of these heat transfer mechanisms. Non-negligible heat transfer would be key for evaluating neutron feedback in all scenarios.

2.9. *Time scale, energy release, and power distribution.* Several references[2, 4, 27] criticize the paper for misinterpreting the time scale and energy generation rate leading to the postulated explosive release of energy, although limited or no quantitative analyses have been performed to substantiate the criticisms. In part, it is argued that inclusion of a system's temporal development during an approach to criticality will affect and possibly limit the violent nature of the critical excursion, and that the energy generation rate of a critical system will not exceed the disassembly rate for reasonable scenarios. As Reference 27 indicates, the paper unrealistically assumes that plutonium vapor moves uniformly outward through rock at its molecular velocity. Desired modelling would be transient in nature and would characterize limitations of Pu vapor transport mechanisms under such conditions (e.g., rock heating, anisotropy, and spacing of fissures). No distinction is made in the paper between explosive yield and total nuclear heat, although yield will be much less than total nuclear heat. The absence of a shock wave will make the hypothesized "explosion" much less violent than an equivalent-yield explosive device.

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