Title
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Journal
Transactions of the American Nuclear Society, 116

ISSN
0003-018X

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Publication Date
2017

Peer reviewed

The Criticality Safety Studies of Joonhong Ahn

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I. INTRODUCTION

The recent passing of UC Berkeley professor Joonhong Ahn at the age of 57 has been a dear loss to the community of nuclear scientists at large. His contributions to studies on advanced fuel cycle development and the societal and ethical aspects of nuclear technology were known around the world and fostered a great reputation. He produced at least fifty peer-reviewed journal papers, authored or contributed to ten books, and organized a considerable number of international meetings.

Ahn's studies led to his eminence in the area of nuclear waste management, where he focused on repository performance assessments along with their impact on society. These assessments included the development of analytical models for nuclide transport phenomena and evaluation of the possibility of criticality events imparting a dose to the biosphere. This paper will summarize his achievements in criticality safety along with developments before his passing.

II. BACKGROUND

Evidence for underground criticality is based on experimental measurements of the uranium deposits in Oklo, Gabon. This "natural reactor" sustained chain reactions two billion years ago and was formed after an oxidative shift in the environment allowed the transport of dissolved uranium species and eventual re-concentration into a critical configuration in reducing sediments.

Using the Oklo analogue as a reference, scientists proposed the need for a criticality safety assessment (CSA) for the direct disposal of used nuclear fuel (UNF) in 1978.[1] Given the large source term of fissile material that would be emplaced in the canisters, it was hypothesized that a critical mass could be formed underground after the breaching of canisters. However, it was determined that the mass of the deposition would have to be much larger than that observed at Oklo given the low enrichment of UNF. This, along with the improbable configuration required to achieve criticality, led to the issue being discarded.

In 1979, the U.S. government had established the Waste Isolation Pilot Plant (WIPP) for the disposal of transuranic (TRU) waste generated from weapons production facilities. A report from the year prior suggested that although short-term criticality events from storage configurations were infeasible, criticality control would be a long-term concern if the thermally fissile material (TFM) were to leach and re-concentrate.[2] While model development was recommended to describe the transport mechanisms involved, the phenomenon was removed from consideration due to the relatively small concentrations of fissile nuclides in TRU waste.

The Department of Energy (DOE) was tasked with the final disposal of high-level waste (HLW) and UNF through the Nuclear Waste Policy Act of 1982. The subsequent amendment in 1987 designated the Yucca Mountain Repository (YMR) as the site of interest. Criticality control standards were established through the 10-CFR-60 regulation of 1983, where criticality events upon emplacement were prohibited by engineering design. However, post-closure criticality was left as part of the total system performance assessment (TSPA) prescribed in 2001 with 10-CFR-63.

The long-term criticality issue gained momentum in the early half of the 1990s with the impending issue of long-term disposal for excess weapons-grade plutonium (w-Pu) to satisfy non-proliferation agreements. This brought about an initiative to evaluate the environmental hazards of potential options for final disposition. As a result, the immobilization of the plutonium stockpile followed by disposal deep underground (along with burnup in existing reactors) was eventually chosen as the solution.

In 1995, a controversial report proposed scenarios in which w-Pu and highly-enriched uranium (HEU) could be naturally reconfigured underground into a supercritical mass with autocatalytic, or self-sustaining, chain reactions.[3] This study analyzed certain water-ejection and ingress scenarios for idealized, homogenous critical configurations of TFM with bedrock and groundwater serving as moderators. It was suggested that for certain cases, given positive reactivity feedback, the lack of a negative feedback mechanism could lead to a substantial release of energy. Criticisms of this study included the incongruence in the time scales for the events involved, the neglect of preventative engineering practices, and inadequate details on the necessary transport processes.[4] Nonetheless, the paper was an impetus for further investigation.

III. BODY OF WORK

III.A. Underground Criticality at Yucca Mountain

In 1995, Ahn began his participation on a project that explored the scenarios in ref. [3] as part of a CSA. The goal was to verify the possibility of autocatalytic criticality from TFM vitrified in borosilicate glass emplaced in the YMR geology. The investigation involved scenario development, the construction of deterministic models to evaluate performance measures, statistical uncertainty evaluations, and remarks on safety with regard to established standards.

III.A.1. Scenario Development

A scenario event tree was created to demonstrate the necessary events for the venting of radioactivity to the biosphere.[5] Given certain moderating conditions and the fissile content of the waste, processes were identified as crucial for the phenomenon to take place: canister degradation, the flushing of neutron poisons, nuclide transport, wide availability of TFM throughout the repository, a common point of precipitation, and positive reactivity feedback mechanisms. As a final step, the effective neutron multiplication factor must be kept above unity in order for a sufficient amount of energy to be generated before negative feedback can have a quenching effect. This, in turn, leads to the venting of radionuclides.

As a direct response to the improbable homogeneous configurations employed in the analysis in ref. [3], a static neutronics analysis was conducted pertaining to TFM depositions in more realistic parallel planar fractures. It was assumed that TFM would deposit uniformly as oxides within the fractures, and that the remaining space would be occupied by water. A reference composition of rhyolite tuff from YMR was used as opposed to pure silicon dioxide to acknowledge site-specific moderation characteristics.

Several reactivity feedback effects were considered independently: water ejection, heating of TFM, heating of the rock, homogenization, thermal expansion, and the buildup of fission products. For implementation in Monte Carlo code, the critical system was modeled either as an infinite lattice of repeating units of rock, TFM, and water, or a homogeneous combination of those components bounded by a spherical reflector with a finite radius.

The undermoderated in-canister criticality case involving w-Pu considered in ref. [3] was dismissed as speculative. In this scenario, soluble poisons would leach from the canister and plutonium would disperse into the dry, moderating surrounding rock to form a critical mass. Heating would eventually cause the metal to vaporize and vent through surrounding fractures, serving as a positive feedback mechanism. In reality, it was realized that unattainably large amounts of TFM would be needed for dry criticality, and that poisons could be selected with low solubilities matching that of plutonium to prevent flushing.

For wet, overmoderated criticality scenarios, the neutron-absorbing aspect of groundwater outweighs its moderating effect. Given a highly-enriched deposit without neutron poisons, it was shown that complete expulsion of water from the fracture could raise infinite multiplication above unity, suggesting that the mixing of TFM and rock would be a matter of concern. When such homogenization is taken to account, this serves as a significant reactivity insertion mechanism by reducing spatial self-shielding. In general, if dry-out is extended to the host rock itself from fission-heating, a positive reactivity feedback mechanism is created and k_∞ increases. However, the effect on the k_{eff} value is much smaller due to neutron leakage effects.

Feedback based on the heating of TFM depends largely on the fissile content. For the host rock, heating causes thermal expansion and spectrum hardening, which have competing effects on reactivity. In a heterogeneous system, it was found that the Doppler effect takes precedence to spectrum hardening due to heat transfer limitations.

Overall, despite the mechanisms and possibility of autocatalytic criticality being demonstrated, such an event was considered unlikely to occur from HLW in the YMR environment. This was based on the high immobility of plutonium and the lack of reducing conditions to allow for the precipitation of uranium solutes, making the configurations and reactivity patterns very unachievable. Furthermore, engineering measures, such as reprocessing, could be undertaken as preventative measures.

III.A.2. Transport Modeling of TFM

Criticality scenarios could not be excluded based on the static neutronics analysis. To provide a robust transport basis for the results of this analysis, Ahn compared a pure-colloid transport model for w-Pu and a pure-solute model for uranium, w-Pu, and boron to verify the extent of nuclide migration in YMR.[6] The goal of the analytical approach was to provide upper and lower bounds to migration distance from vitrified plutonium waste in a fractured system. A glass matrix with a set source term of TFM was assumed to dissolve in groundwater, upon which individual species would undergo a solubility-limited release into fractures. For the colloid model, Pu colloids were not subject to rock matrix infiltration and were assumed to be governed by a flocculation process. Boron was assumed to be readily soluble and its sorption effects negligible.

It was discovered that plutonium can accumulate in large amounts close to the fracture entrance via colloid-based transport and that significant migration can be observed on the order of several meters. However, the total accumulated mass would be too small to sustain criticality. For solute-based transport, a small amount of Pu remains in proximity to the repository and decays to negligible amounts. Uranium spreads much further down the fracture due to greater mobility and re-entry from sorbed species in the rock matrix. However, even with the flushing of boron poison, U concentration remains below the critical threshold. It was concluded that a more sophisticated plutonium transport model combining solute and colloid mechanisms was necessary, along with intermittent water flows for unsaturated geology.

III.B. Granitic Repository Concept

III.B.1. Criticality Safety Assessment

A CSA was undertaken for HLW from the reprocessing of commercial light water reactors (LWR) emplaced in a granitic geology in 1997.[7] It employed a solute transport model for parallel planar fractures in contact with the engineered barrier systems (EBS) of a water-saturated repository, which was implemented in the Transport to Biosphere computer code.[8] Conservatism was based on the overestimation of TFM. leading to an assumption that contributions from each canister would contribute to a common point of accumulation in the far-field without mass-transfer limitations posed by adjacent canisters or steel overpack.

Based on transport parameter variation, cases were proposed based on the arrival times of nuclides to the surface of the EBS. Using a set source term, total precipitation was calculated at various points in the far-field. The case promoting uranium arrival to the EBS surface was observed to be most conducive to extending the uranium plume into the far-field and maximizing enrichment. Verification of the minimum critical mass followed for enrichment figure obtained from those results; that is, if the minimum critical mass was too large, it would be possible to exclude the criticality event.

Using a reference granite composition, it was assumed that uranium accumulated as UO_2 in saturated rock of 10% porosity in a homogenized reflected sphere configuration. The minimum critical mass and core radius were found to be much higher for granite than pure SiO_2 because of greater absorption properties. The U mass requirement for granite seemed unachievable in the suggested spherical volume. However, when the water content of the system was increased to 30%, the minimum critical mass was found to dramatically decrease.

With the absolute minimum critical masses at hand, it was necessary to analyze the minimum mass needed to be *overmoderated* as a precedent for autocatalytic criticality. For granite, given positive reactivity feedback, this mass would not actually result in autocatalytic criticality for a homogeneous system. It was therefore suggested that adding depleted uranium (DU) to the HLW packages would be a suitable measure to reduce the enrichment of the far-field accumulation and raise the necessary critical mass for self-sustaining chain reactions. Furthermore, it was suggested that efficient minor actinide recovery from the reprocessing of UNF could enhance criticality safety.

III.B.2. Analysis of Uncertainty

A simplified transport model was devised in 1998 that neglected the role of precursors. Individual waste forms and their surrounding buffers were modeled as sequential compartments interacting with each other and the host rock through diffusion and advection. Inter-canister effects on concentration were included as a measure to reduce conservatism in the model. A further analysis was made to probe the uncertainty of the maximum mass of U-235 in the far-field.[9]

Distributions were assumed for key transport variables and system properties, while other parameters remained fixed. Samples from the distributions were obtained for several different cases via the Latin hypercube sampling method, which were then applied to the deterministic model for a set source term. It was found that acknowledging concentration limitations between sequential canisters markedly decreases the release of uranium from waste downstream of the direction of groundwater flow.

Although the amount of U-235 depositing in the far-field would be enough to account for autocatalytic criticality, it was maintained that the event was unlikely because of the need for all uranium to accumulate at one single location with the specific geometry requirements. In reality, depositions would take place in a heterogeneous geometry with a much higher threshold for criticality. Although a transport basis was established for granite, site-specific studies were still recommended.

III.B.3. Review

As part of an integrated repository-wide TSPA, a preliminary CSA was conducted in accordance with the Japanese repository concept for a fractured, water-saturated granitic host medium for HLW from the reprocessing of UNF.[10] The fracture model of ref. [8] was employed for bounding mobility cases of uranium and its precursors. It was found that the mass of uranium required for over-moderated criticality in low-porosity granite is too massive to make an autocatalytic event a concern. U-235 was specified as the TFM of interest, with other fissile actinides considered to be of less concern due to short half-lives and packaged poisons. A colloid-facilitated transport analysis was suggested for consideration once site-specific details became available.

A comprehensive review paper was written in 2006,[11] where it was acknowledged that the necessary conditions for criticality and positive feedback are, in fact, conceivable, yet unlikely. For YMR, it was stated that the transport behavior of both plutonium and uranium make critical configurations largely unobtainable. For the water-saturated granitic repository, results were inconclusive but implied that autocatalytic criticality events were still highly improbable.

Engineering measures were proposed to further reduce the probability of autocatalytic criticality while meeting licensing requirements. This included the use of DU or insoluble gadolinium as opposed to boron as a packaged poison, and the implementation of advanced fuel cycles that eliminate uranium precursors. Suggestions were made for the characteristics of a candidate site, including low-porosity bedrock and a low concentration of reducing agents in the geochemical environment.

Important differences were highlighted between a CSA and a performance assessment based on radiological hazard to the environment. Conservatism for a CSA concerns the overestimation of TFM deposition in the bedrock, which could be achieved by promoting higher nuclide sorption characteristics in transport modeling. By contrast, performance assessments based on radiological hazard try to overestimate the concentration of nuclides in downstream groundwater that result in potentially higher doses to the biosphere. Furthermore, although reduction in toxic radionuclides in canisters could enhance criticality safety, such measures would be of limited efficacy to reduce total hazard. Overall, a TSPA would need to acknowledge both concerns along with other considerations such as proliferation resistance.

III.C. Fukushima

The Fukushima Daiichi (FD) accident of March 2011 prompted studies on the final disposition of nearly 250 MT of damaged fuel from the affected reactors. Although the characterization of the fuel had yet to take place, it was assumed that final disposal in a deep geological repository would eventually be required. As part of his influential response to the tragedy, Ahn supervised a CSA for a repository containing the damaged UNF from the three affected reactors.[12] This was a parametric study that expanded the static neutronics analyses begun in ref. [5] by systematically evaluating the effect of heterogeneous critical configurations. The fuel composition was obtained from burnup calculations based on the FD reactor operation schedule and an assumed interim cooling period.

The criticality scenario was based on the accumulation of U and Pu oxide in the far-field for both a homogenous agglomeration and a finite system of saturated planar fractures. The neutronics analysis was parameterized by varying proportions of metal and water in the void space of either of two rock compositions. The fracture width was also varied to examine its effect on neutron multiplication and the minimum void fraction required for a critical mass.

It was demonstrated that various far-field critical configurations are possible based on a range of material conditions and geological formations. For the fractured geometry, numerical exploration demonstrated the impact of spatial self-shielding. To further the evolution of the static neutronics analysis, it was suggested that randomized fracture geometries and volumes be studied in the future.

IV. CLOSING REMARKS

Although YMR was moth-balled in 2011, Ahn's studies provided important context to the criticality issue and established invaluable principles and methodology for site-specific studies. His particular approach to CSA is still being emulated in current research for final waste disposition in Japan. Before his passing, studies were underway on nuclide transport modeling for UNF and FD damaged fuel in a granitic repository, along with more extensive parametric studies for the neutronics evaluation. Novel in-canister criticality studies were completed as part of the last doctoral dissertations under his complete supervision.

Continuing studies on the criticality issue at Berkeley are largely based on the future work suggested in Ahn's literature and direct guidance given before his passing. Although these studies are inconclusive on verifying the criticality phenomenon from direct disposal in granitic geology, it has been maintained that the phenomenon cannot be excluded outright, and that the issue still merits attention for repository performance.

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