FOR:	The Commissioners
FROM:	L. Joseph Callan /s/ Executive Director for Operations
SUBJECT:	POST-DISPOSAL CRITICALITY RESEARCH

#### PURPOSE:

To inform the Commission of the staff's assessment of whether to continue post-disposal criticality research at low-level waste (LLW) disposal facilities, and to obtain the Commission's direction on additional research.

#### BACKGROUND:

This paper responds to the April 29, 1998, Staff Requirements Memorandum (SRM) concerning SECY-98-010, "Petition for Envirocare of Utah, Inc., to Possess SNM in Excess of Current Regulatory Limits" (Attachment 1). This SRM directed the staff to review the Oak Ridge National Laboratory (ORNL) report on post-disposal criticality at the Barnwell, South Carolina, disposal facility and to inform the Commission of its findings and recommendation on whether to continue post-disposal criticality research. The SRM also directed the staff to consult with the Advisory Committee on Nuclear Waste (ACNW) on generic issues.

As part of the staff's evaluation of the petition for rulemaking submitted by Envirocare of Utah, Inc. (Envirocare), staff performed a bounding analysis to evaluate the potential for special nuclear material (SNM) to migrate and reconfigure at LLW disposal facilities, resulting in an inadvertent criticality. As a result of this bounding analysis, the staff concluded that post-disposal criticality concerns could not be dismissed as a possibility. Given the uncertainties associated with the assumptions and scenarios evaluated, the staff determined that technical assistance was required to further evaluate the hydro-geochemical processes necessary to result in a post-disposal criticality. ORNL was contracted in 1995 to provide this technical assistance, using the Envirocare site as a model. Following the study at Envirocare, the staff concluded that a LLW disposal site, in a humid climate, using different disposal methods, should be evaluated to better determine the likelihood of post-disposal criticality at LLW disposal facilities. The Barnwell disposal facility was selected as a model for this additional study.

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In 1997, the staff identified the need for research in two areas of LLW criticality. The first research need was prompted by the Commission's direction in the SRM on SECY-96-268, "Final Rule to Amend 10 CFR Part 71 for Fissile Material Shipments and Exemptions" (Attachment 2). The revisions to Part 71 set limits on unusual moderators such as beryllium, when shipping fissile material. Babcock and Wilcox (B&W) identified this concern during shipment preparation of a waste product from the down-blending of weapon-usable fissile material that could result in nuclear criticality. The Commission directed staff to consider the criticality issues raised in SECY-96-268 in a broad context and to examine previously unanticipated fissile materials and moderators in other areas of the fuel cycle and waste programs. The second area of research was to develop a generic methodology to quantify the risk associated with post-disposal criticality. These two issues were consolidated, and a statement of work (SOW) was sent to ORNL. In response to the SOW, ORNL submitted a cost proposal on March 29, 1998. This research was placed on hold pending implementation of the Commission's direction in the SRM on SECY-98-010.

Consistent with the SRM on SECY-98-010, the staff briefed the ACNW on July 20, 1998. The presentation described the previous studies (i.e., ORNL's studies of Envirocare and Barnwell and the staff's work in support of the "Draft Environmental Impact Statement for the Shallow Land Disposal Area in Parks Township, PA"), and discussed the limitations of these studies in the context of the research requests. In response to this presentation, ACNW issued a letter dated July 30, 1998, concluding that significant research on post-disposal criticality was not warranted. The Committee stated that the studies contained elements of a risk assessment but lacked consistency of application in propagation of realistic uncertainties through the analytical model. It recommended performing a quantitative risk assessment on a specific site and that this assessment be externally peer-reviewed.

# DISCUSSION:

The staff's evaluations of the Envirocare, Barnwell, and Parks Township studies are discussed in Attachment 3. This evaluation included performing additional hydrologic and geochemical evaluations for the Barnwell site to determine whether additional research should be conducted. A copy of the Barnwell study is included as Attachment 4. As discussed in Attachment 3, if the trench cover remains in place, under reasonably credible scenarios, a post-disposal criticality would be unlikely for 10,000 years. Staff also did an analysis conservatively assuming the trench cover is removed after 500 years, and found that criticality was unlikely for 1000 years. The consequences of a post-disposal criticality were not evaluated for the Barnwell site. Although the uncertainties associated with this likelihood estimate have not been completely quantified, the studies performed to date indicate that, while theoretically possible, post-disposal criticality is unlikely. Based on the additional work, staff concludes that additional research is not a high priority and that the uncertainty does not need to be precisely quantified. However, additional work could be performed if the Commission wishes to quantify this uncertainty.

Attachment 5 is a differing professional view (DPV; as allowed under Management Directive (MD) 10.159) on the conclusions in this Commission Paper, submitted by a staff member on October 2, 1998. The staff will review the DPV in accordance with the procedures in MD 10.159.

With respect to the unusual moderator issue raised in SECY-96-268, the staff responded to the SRM on SECY-96-268 in a memorandum to the Commission, dated May 21, 1997. Staff concluded that the fuel cycle regulatory process is adequate to ensure public safety with respect to criticality issues of unanticipated moderators and fissile material. Staff proposed evaluating whether or not there is a need to restrict or provide constraints on the burial of unusual moderators in LLW facilities. As discussed above, the first research need noted that unusual moderators had not been considered in the previous post-disposal criticality studies. Staff recommended this research in order to reduce any residual uncertainty concerning the issue of unusual moderators in waste.

Unusual moderators as they pertain to emplacement LLW criticality have been evaluated in NUREG/CR-6284, "Criticality Safety Criteria for License Review of Low-Level Waste Facilities." This report states that mass of beryllium should be limited to five times the mass of uranium-235 (U-235) and the mass of carbon (graphite) should be limited to twenty times the mass of U-235. To gain a general sense of the presence of unusual moderators in LLW, staff contacted State officials, familiar with past disposal at LLW sites. Staff was informed that, during the past several years, large quantities of unusual moderators (beryllium and graphite) have not been disposed of at the Barnwell, Richland, and Clive sites, although the presence of such unusual moderators is not routinely noted on LLW manifests. This is consistent with staff's knowledge of LLW disposal practices. With the exception of B&W, staff is not aware of any significant sources of beryllium among facilities licensed by NRC. In addition, large sources of graphite would be more economically disposed of by incineration. Further, 10 CFR Part 71 limits the quantity of unusual moderators in fissile exempt shipments well below the limits identified in NUREG/CR-6284. Even if significant quantities of unusual moderators were present in LLW, it is unlikely that they would be commingled with SNM to create a critical array.

Staff concludes that significant sources of unusual moderators are not present at LLW facilities. In addition, in response to the SRM on SECY-98-010, staff is preparing guidance on emplacement criticality and will emphasize the need to limit significant quantities of unusual moderators in LLW disposal units, which is consistent with existing practices.

#### Options:

The staff has identified the following three options on post-disposal criticality research. If option 1 or 2 is selected, staff would not continue with further evaluation of unusual moderators. For the reasons cited above, staff considers that research on unusual moderators in LLW disposal independent of research on post-disposal criticality is not warranted. If option 3 is selected, unusual moderators would be included as part of the generic methodology.

1. Cease further review of post-disposal criticality. Existing studies indicate that long-time frames would be required to reconfigure the SNM into a critical mass. Monitoring of wells and sumps at disposal facilities, as suggested by ORNL, could mitigate any remaining concerns by providing early warning of any sign of reconcentration in the trenches. However, signs of problematic conditions, if they occur, would be expected to take hundreds of years to appear and reliance on long-term monitoring is not consistent with the institutional control provisions in 10 CFR 61.59.

The principal advantage of this option is that no additional resources are required. Option 1 does not provide a comprehensive basis for concluding that post-disposal criticality poses no significant health and safety concern, because the uncertainties have not been fully quantified. Staff's analyses suggest that the likelihood that criticality will occur is very low over thousands of years.

2. Conduct a limited-scope study to reasonably quantify the associated risk. This is the approach recommended by the ACNW. To implement this approach, staff would request technical assistance to evaluate an additional two to three trenches at the Barnwell site, using a two-dimensional or three-dimensional flow and transport computer code to model radionuclide transport under variably saturated conditions. Probability density functions would be established for key model parameters to evaluate the uncertainty. A probabilistic approach would be used to calculate the maximum U-235 accumulation and quantify the likelihood of occurrence. The consequences for any potential criticalities would be determined. The risk would be obtained by multiplying the likelihood times the estimated consequences. If this option is selected, staff would brief the ACNW on this scope and obtain the Committee's input before proceeding with the technical assistance.

The principal advantage of this option is that it may reasonably quantify the risk of a post-disposal criticality with fewer resources than option 3. The results from this effort might provide the staff with a firmer basis for concluding that post-disposal criticality is not a significant concern. However, this option would not develop a generic methodology that could be used to evaluate other sites.

3. Develop a generic methodology. This option would develop a methodology, to quantify risk from a post-disposal criticality, that could be used by existing and future LLW disposal facilities. This option would include consideration of unusual moderators in LLW disposal.

The principal advantage of this option is that it produces a tool which could be used in the future to quantify the risk associated with post-disposal criticality. However, expending the resources to complete this option may not be warranted, considering the perceived small risk.

#### **RESOURCES**:

Option 1 would not require any resources. The staff estimates that the cost to complete option 2 is approximately \$250K in contractor support and approximately 0.3 full-time equivalent (FTE). The staff estimates that the costs to complete option 3 is approximately \$400K in contractor support and approximately 0.5 FTE. Resources to conduct the activities described in these options are currently not included in the FY 1999-2000 budget for the Office of Nuclear Regulatory Research or the Office of Nuclear Material Safety and Safeguards. If option 2 or option 3 is chosen, resources will be reviewed during the next budget cycle.

# **RECOMMENDATIONS:**

Considering the current staff workload, budget constraints, and the apparent low likelihood of reconcentration and criticality, the staff considers that

additional technical assistance and/or research is of low priority, and therefore, recommends option 1, cease further review of post-disposal criticality. The staff considers this option to be consistent with the memorandum from the Chairman to the Executive Director for Operations, dated August 7, 1998, which instructed staff to prioritize areas and increase the threshold for commencing new initiatives.

## COORDINATION:

The Office of the General Counsel has reviewed this Commission Paper and has no legal objections. The Office of the Chief Financial Officer has reviewed this paper for resource implications and also has no objections.

L. Joseph Callan Executive Director for Operations

Attachments: As stated

ATTACHMENT 1

SRM on SECY-98-010

ATTACHMENT 2

SRM on SECY-96-268

ATTACHMENT 3

# DWM STAFF EVALUATION OF PREVIOUS POST-DISPOSAL CRITICALITY STUDIES

- A. INTRODUCTION
- B. SUMMARY OF STAFF ANALYSES FOR PARKS TOWNSHIP
- C. SUMMARY OF ORNL ANALYSES OF ENVIROCARE AND BARNWELL
- D. CONCLUSIONS
- References

#### A. INTRODUCTION

The U.S. Nuclear Regulatory Commission has evaluated three low-level waste (LLW) and decommissioning sites for post-disposal criticality: (1) Parks Township, Pennsylvania; (2) Envirocare, Utah; and (3) Barnwell, South Carolina. NRC staff evaluated the Parks Township site with some assistance from Oak Ridge National Laboratory (ORNL). ORNL evaluated the Envirocare and Barnwell sites with some assistance from the staff. These three sites have several attributes in common:

- 1. They disposed of uranium, including depleted uranium (DU) and special nuclear material (SNM) in the form of enriched uranium.
- 2. There may be sufficient inventory and enrichments of uranium to lead to a theoretical criticality under ideal circumstances.
- 3. The enriched uranium was widely dispersed with soil or other wastes and often with much-larger quantities of DU or natural uranium.

## **B. SUMMARY OF STAFF ANALYSES FOR PARKS TOWNSHIP**

The Parks Township Shallow Land Disposal Area received waste, including SNM from the Apollo plant (NRC, 1997). The staff analyzed the potential mechanisms of SNM migration from a dispersed state to a concentrated state, with favorable geometry and water content that could lead to criticality. The criticality calculations assumed uranium with an enrichment of 20 percent. The analyses were limited to 15,000 years, judged to be the maximum time that the trenches would remain intact before erosional forces would disperse their contents. A critical configuration would require the migration of uranium from a highly dispersed state to a concentrated state. Vertical migration of uranium would not lead to criticality. Focused flow toward a hypothetical drain with capture of virtually all uranium would be required to accumulate a potential critical mass. However, the staff concluded that the accumulation of a critical mass within a single trench would require a highly efficient concentration mechanism to operate at the expense of other, more likely redistribution processes. Using the available data, the analyses indicated that there was reasonable assurance, making allowances for the time period and uncertainties involved, that the potential for a critical mass to form was so unlikely that criticality need not be considered further. It should be noted that the SNM inventory at the Parks Township site is significantly less than at LLW disposal sites.

## C. SUMMARY OF ORNL ANALYSES OF ENVIROCARE AND BARNWELL

The Envirocare study examined mobilization, migration, and concentration of uranium initially dispersed in soil, using geochemical transport models (ORNL, 1997). This study considered the two concentration processes: (1) sorption onto materials such as iron oxide; and (2) precipitation in a chemically reducing zone. The analyses demonstrated that nuclear criticality could occur under certain unlikely conditions by vertical reconcentration of the uranium to a thinner layer at the bottom of the disposal cell, by the process of chemical reduction and precipitation. Sorption processes alone did not lead to any critical conditions for the cases studied. This study assumed that the source was 100 percent enriched uranium at the maximum allowable concentration specified in the State of Utah's license. These conditions do not exist at the Envirocare site, and the actual conditions are much less

conducive to nuclear criticality. The estimated average enrichment of the inventory through 1993 is only 0.42 percent, which is less than natural uranium and the theoretical minimum enrichment necessary for criticality in a water-moderated system.

The Barnwell study examined precipitation of uranium in reducing zones postulated to be caused by decaying organic material such as wood and cardboard, and corroding iron such as steel drums. However, the study noted that reducing zones would require saturated conditions, which do not presently exist in the trenches at the site (ORNL, 1998). This analysis assumed that the uranium was enriched to either 10 percent or 100 percent and initially in containers. Unlike the Envirocare evaluation, the Barnwell study considered both vertical migration of uranium to the bottom of the trench, and then horizontal transport, within the trench, to form a critical geometry of reconcentrated material.

The Barnwell study concluded that the minimum concentration of uranium-235 (U-235) to produce a criticality was 1.6 times greater for 10 percent enrichment than for 100 percent enrichment. Below 10 percent enrichment, significantly greater concentrations of U-235 are required. Assuming the formation of reducing zones under saturated conditions (limiting oxygen in the system), reducing zones may be effective in precipitating uranium and appear to be stable even with the influx of oxidizing water. However, to date, the formation of reducing zones is hypothetical and has not been observed at Barnwell. For the conditions assumed in the Barnwell report, approximately 10,000 years would be required to mobilize and reconfigure the uranium. Based on actual uranium areal density of the disposal trenches, horizontal flow would be required to increase the areal density to pose a criticality concern.

ORNL suggests monitoring the sumps at the site for uranium, iron, and organics. If uranium is detected, its enrichment should be determined, and the redox condition should be evaluated by determining the speciation of iron. ORNL notes that placing caps over the trenches will limit infiltration and promote oxidizing conditions. Commingling SNM and source material (natural and depleted uranium) will reduce the average enrichment and thus reduce the concern of post-disposal criticality. Chem-Nuclear LLC, operator of the Barnwell facility, has been constructing caps over the waste trenches as part of the tritium migration mitigation.

#### 1. Simplifying assumptions and uncertainties

Some of the simplifying assumptions used in the Envirocare and Barnwell analyses included: (1) uniform uranium distribution in waste; (2) onedimensional flow; (3) saturated conditions; (4) stable reducing zones; and (5) U-235 and U-238 mobilized uniformly. The reports concluded that criticality might be possible under certain unlikely conditions, and long-time frames would be required. Assuming average enrichment and areal density, post-disposal criticality does not appear to be likely at the Envirocare and Barnwell facilities.

One key uncertainty with the ORNL studies is the assumption that the enrichment and U-235 concentration are homogeneous throughout the trench. For trenches 1 to 36, only the quantities of enriched uranium were reported, so it was conservatively assumed in the criticality evaluations that the inventories were 100 percent enriched in U-235. Trenches 37 and above reported both SNM and source material. These records clearly show that large amounts of source material are commingled with the SNM in most trenches, or, in one case, that the amount of disposed SNM was very small. In the Barnwell study, one of the older trenches, for which only SNM was reported, (trench 23) was identified as being of possible concern because of its high mass of U-235. To resolve this concern, the staff obtained trench loading records from the State of South Carolina. ORNL analyzed these records and concluded that there is three orders of magnitude more source material than SNM, and that the materials are commingled sufficiently that the assumption of uniform enrichment is reasonable. However, other trenches might contain localized areas of concentrated and enriched uranium that could pose a potential concern. Localized concentrations of SNM in areas relatively free of source material could lead to accumulation of a critical mass in shorter time frames if there is "funneling" or convergence of flow to a central location; immobilization mechanisms for uranium that worked efficiently; and configuration of the accumulated mass that had favorable neutronic properties. The staff cannot completely rule out this possibility without evaluating detailed disposal records, such as was done for trench 23.

Another area of uncertainty that may be mitigative is that the studies did not consider the effects of kinetics of sorption and redox reactions. Some research suggests that precipitation of uranium does not occur readily at low temperature (Meunier, et. al, 1990; Duff, et. al, 1997). Further, the ORNL studies assumed reducing conditions would exist and be stable for time frames necessary to reconcentrate the SNM, which has not been demonstrated, and in fact is contraindicated in most landfills (e.g., Greenfield, et. al, 1990). Other assumptions that were not considered and could affect the time required to reconcentrate SNM include: (1) transient infiltration and concentrated flow conditions; (2) variable container weathering; and (3) combination or competition of various geochemical processes.

#### 2. Additional analyses at Barnwell

Staff conducted additional analyses for the Barnwell site to determine whether the Agency needs additional research and technical assistance regarding criticality at LLW disposal and decommissioning sites. The staff performed a computer modeling study of flow in the trench under the influence of infiltration at the surface, with and without the soil cap in place. To simulate moisture movement in a typical Barnwell trench, the staff used a two-dimensional, variably saturated flow code (Celia, 1990). The trench was assumed to be 23 meters wide, six meters deep on one side and sloping to seven meters deep on the other side. A one meter deep French drain was modeled on the bottom of the lower side of the trench. The water table was assumed to be five meters below the trench bottom. Recharge in the native soil around the trench was treated as a steady state, constant flux boundary equal to  $1.2 \times 10^{-6}$  cm/s along the top of the model.

The condition of a failed soil cap was treated as a constant flux boundary equal to 3.8 x 10<sup>-6</sup> cm/s to conservatively represent no cover over the trench.

The staff used a value of hydraulic conductivity in the clayey sand of  $10^{-5}$  cm/s. The modeling exercise with the missing cap showed that a moisture front reaches the trench bottom within the first year. There appears to be some lateral flow into the French drain, but no ponding there because water exfiltrates faster than it can accumulate.

Staff attempted to model moisture movement, assuming a lower hydraulic conductivity of  $1.4 \times 10^{-7}$  cm/s for the clayey sand, but was unable to obtain convergence with the computer code. The staff believes that the higher values are more reasonably applied to the trench-scale model than the lower reported values, which were derived from small core samples. The higher value of hydraulic conductivity is also consistent with a number of field-scale studies at and near the site.

The staff also performed additional analyses using HELP (Schroader et. al, 1988), a quasi-two dimensional water routing code for analyzing water movement in covers. The staff simulated the as-built trench, and concluded that infiltrating water would flow mostly in the vertical direction, with very little diversion (0.3 m<sup>3</sup>/yr) from the trench to the French drain. The staff's analysis also showed that assuming no soil cap results in a small amount of lateral drainage to the French drain (roughly 5 m<sup>3</sup>/year on average). This analysis also showed that the maximum head on the clayey sand layer would be about 0.15 m, and the peak flow to the French drain would be 2 m<sup>3</sup>/d, leading to the conclusion that there would be little if any ponding within the trench. These analyses show that with a soil cap on the trench, the time needed to accumulate a sufficient mass of U-235 is on the order of 10,000 years, as predicted by ORNL. Staff also did an analysis in which it conservatively assumed that the trench cap was removed at 500 years and determined that at least another 500 years would be needed to reconfigure the SNM to form a critical mass. These time estimates assume that the reconfigured uranium has an enrichment of 10 percent. However, disposal records indicate that the average enrichment is significantly lower, and in most cases, below the minimum 1 percent enrichment needed to produce a criticality. Therefore, longer timeframes would be required if the enrichment was between one and ten percent. Moreover, there are additional reasons, which are discussed below, why post-disposal criticality is not likely.

Because it had been assumed that reducing conditions would be required to precipitate uranium, the staff also performed additional geochemical calculations, using PHREEQC (Parkhurst, 1995), to determine if uranium could be reconcentrated under oxidizing conditions (i.e., the conditions expected to exist in the Barnwell trenches). The staff modeled the exposure of urananite to water under oxidizing conditions. Such exposure resulted in the conversion of urananite to schoepite (another species of uranium oxide). This solution in a medium containing silica (i.e., sand such as exists in the Barnwell trenches) resulted in the precipitation of soddyite (another species of uranium oxide). The uranium concentration in the solution was significantly decreased, thus immobilizing most of the uranium in solid form. Based on this analysis, the staff concludes that reducing conditions are not required to precipitate uranium. However, the kinetics of these reactions are currently unknown, and so the efficiency of the analyzed mechanism under oxidizing conditions is uncertain.

In summary, there are several lines of reasoning why the staff believes criticality at Barnwell is not likely:

- Actual areal densities of uranium in the trenches requires horizontal flow to accumulate sufficient mass of uranium to form a criticality. With the cover intact, virtually all flow will be vertical in the unsaturated zone, with little if any flow to the French drain. With the cover removed, most flow would still be vertical, but there would be greater diversion, up to 5 m<sup>3</sup>/year, to the French drain. There would not likely be any accumulation in the French drain, however, because water would exfiltrate through the clayey sand as fast as it infiltrated.
- Long-term ponding of water to form stable reducing conditions does not appear likely. Because the French drain will be above the water table and generally well-drained, even with the soil cap removed, the environment should be oxidizing, thereby promoting the rapid decay of organic matter. Being near the bottom of the trench, there would be no likely avenues for organic material from the surface (e.g., dead leaves) to accumulate. Even with the cover in place, the sampled groundwater in the trenches is moderately oxidizing.
- Some scientific evidence in the literature indicates that urananite does not form quickly or easily at low temperatures, even in highly reducing zones (e.g., Meunier, 1990; Duff, 1997). However, precipitation of uranium in oxidizing conditions is possible (e.g., Hemingway 1982, Moll et. al, 1996, and Nguyen et. al, 1992). Even if it can be assumed that precipitation of uranium occurs in oxidizing conditions, the uranium would become essentially immobile in the drainage sand layer, and increased concentration of uranium due to horizontal flow would not occur.
- The average enrichment of the uranium in the trenches is likely to be low. Trenches for which the inventories of SNM and source material were reported generally had low average enrichments or very small inventories of SNM. There remains uncertainty about the average enrichment in the older trenches, for which only SNM quantities were reported. However, one of those trenches, trench 23, had less than 0.3 percent average enrichment when monthly disposal records were inspected. Although there might be local pockets of higher enrichment in any of the trenches, the scenario that depends on migration of uranium from a large area to the French drain would also homogenize the enrichments to a uniformly lower level.

#### D. CONCLUSIONS

The NRC staff and its contractors have completed three evaluations of potential in-ground criticality at LLW disposal and decommissioning sites where enriched uranium was disposed. Although the uncertainties have not been fully quantified, these studies conclude that post-disposal criticality, while theoretically possible, is remote or unlikely.

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ATTACHMENT 4

# NUREG/CR-6505, Vol. 2, "THE POTENTIAL FOR CRITICALITY FOLLOWING DISPOSAL OF URANIUM AT LOW-LEVEL WASTE FACILITIES"

ATTACHMENT 5

DIFFERING PROFESSIONAL VIEW