

EVALUATION OF CRITERIA FOR  
SELECTING A SALT PROCESSING  
ALTERNATIVE FOR HIGH-LEVEL WASTE  
AT THE SAVANNAH RIVER SITE:  
INTERIM REPORT

Committee on Radionuclide Separation Processes for  
High-Level Waste at the Savannah River Site

Board on Radioactive Waste Management  
Board on Chemical Sciences and Technology  
Division on Earth and Life Studies  
National Research Council

NATIONAL ACADEMY PRESS  
Washington, D.C.

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SYBIL A. PAIGE, Administrative Associate

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This report has been reviewed in draft form by individuals chosen for their diverse perspectives and technical expertise, in accordance with procedures approved by the National Research Council (NRC) Report Review Committee. The purpose of this independent review is to provide candid and critical comments that will assist the institution in making the published report as sound as possible and to ensure that the report meets institutional standards for objectivity, evidence, and responsiveness to the study charge. The review comments and draft manuscript remain confidential to protect the integrity of the deliberative process. We wish to thank the following individuals for their participation in the review of this report:

Robert M. Bernero, U.S. Nuclear Regulatory Commission (retired)

J. Brent Hiskey, University of Arizona

Lawrence Kershner, Dow Chemical Company

James W. Mitchell, Bell Labs/Lucent Technologies

Kenneth N. Raymond, University of California, Berkeley

Edwin L. Zebroski, Elgis Consulting

Although the reviewers listed above have provided many constructive comments and suggestions, they were not asked to endorse the conclusions or recommendations nor did they see the final draft of the report before its release. The review of this report was overseen by Royce W. Murray, University of North Carolina, appointed by the NRC's Report Review Committee, who was responsible for making certain that an independent examination of this report was carried out in accordance with institutional procedures and that all review comments were carefully considered. Responsibility for the final content of this report rests entirely with the authoring committee and the institution.





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## SUMMARY

At the request of the U.S. Department of Energy (DOE), the National Research Council formed a committee in 1999 to provide an independent technical review of alternatives selected by the Savannah River Site (SRS) for processing the high-level radioactive waste (HLW) salt solutions stored there. The final report of that committee, *Alternatives for High-Level Waste Salt Processing at the Savannah River Site*, was issued in August 2000. DOE subsequently asked the National Research Council to provide an assessment of DOE's efforts to select a processing alternative for removal of cesium, strontium, and actinides from high level waste at the Savannah River Site. A new committee was appointed, and it addresses in this interim report the first part of its statement of task—"evaluate the adequacy of the criteria that will be used by DOE to select from among the candidate processes under consideration."

DOE identified eleven criteria to be used in evaluating three alternatives for processing the HLW in the SRS tanks. Based on information presented by representatives from the SRS, the committee concludes that the eleven criteria are reasonable and appropriate and were developed in a transparent way. However, as described in the body of the report, some of the criteria do not appear to be independent of others, and some criteria appear unlikely to discriminate among the process alternatives.

The methodology for using the evaluation criteria is still evolving, and revisions in the weighting factors may be necessary in consideration of the points raised in the body of this report. Preliminary application of the criteria in three different scoring exercises by DOE has shown little discrimination among the three processes. The committee recommends that the criteria should not be implemented in a way that relies on a single numerical "total score." Rather than averaging and totaling the scores for each criterion, the various criteria should be seen as relevant to different goals and purposes and should be considered individually. Some of the criteria should be used as "go/no go" gates and some should have thresholds for use that demonstrate a given level of difference between the three processes. Also, the committee recommends that DOE should define what are significant differences in the scoring procedure. The committee finds it difficult to see a path forward for this procedure (e.g., adjustment of weighting factors) without these differences being specified. The objective of the evaluation procedure should be to provide adequate information for making a risk-informed decision evaluating the science, technology, operational aspects, time factors, and costs, as well as policy matters not addressed in this evaluation.

Despite limitations in discriminating among the alternatives, the committee recognizes that research and development currently being conducted for the several alternative processes may result in changes in the scores on the eleven criteria. Additionally, the committee finds that the current scoring system for individual criteria can be useful for identifying and following the progress of the research and development program prior to downselection (i.e., a reduction in the number of process alternatives), thereby assisting in determination of where significant further effort is needed for each process.

## INTRODUCTION

The National Research Council (NRC) formed a committee, at the request of the U.S. Department of Energy (DOE), to provide an independent technical review of alternatives

selected by representatives at the Savannah River Site for processing the high-level radioactive waste salt solutions stored there in tanks. The work of that committee was completed and its findings were reported in *Alternatives for High-Level Waste Salt Processing at the Savannah River Site* (National Research Council, 2000).

After receiving that report, DOE asked the NRC to provide additional advice on the waste processing efforts at the SRS, and a new committee was impaneled to examine the DOE's selection of a process for separating radionuclides from soluble high-level radioactive waste at that site. This newly constituted committee consists of six members of the previous committee, plus three new members whose areas of expertise were needed to address the new charge. The committee was charged with a three-part task:

- 1) evaluate the adequacy of the criteria that will be used by DOE to select from among the candidate processes under consideration;
- 2) evaluate the progress and results of the research and development work that is undertaken on these candidate processes; and
- 3) assess whether the technical uncertainties have been sufficiently resolved to proceed with downsizing the list of candidate processes.

The committee may, at its discretion, also provide comments on the implementation of the selected process. The purpose of this brief interim report is to address the first of the three tasks.

## BACKGROUND

At present three alternative processes remain under consideration for removal of cesium, strontium, and actinides from tank supernate solutions at SRS; namely, small tank precipitation by tetraphenylborate (TPB), ion exchange on crystalline silicotitanate, and caustic side solvent extraction. A brief description of the site's high-level waste program, described in *Alternatives for High-Level Waste Salt Processing at the Savannah River Site* (National Research Council, 2000), is included in this report as [Appendix C](#). A key recommendation in that report was the following:

*The committee finds that there are potential barriers to implementation of all of the alternative processing options. The committee recommends that Savannah River proceed with a carefully planned and managed research and development (R&D) program for three of the four alternative processing options (small tank precipitation using TPB, crystalline silicotitanate ion exchange, and caustic side solvent extraction, each including monosodium titanate processing for removing strontium and actinides) until enough information is available to make a more defensible and transparent downselection decision. The budget for this R&D should be small relative to the total cost of the processing program, but this investment will be invaluable to overcoming many of the present uncertainties discussed in this report.*

Since that report was issued in August 2000, DOE has funded research and development on the three alternative processes, and significant progress has been made in ameliorating many of the technical uncertainties. DOE noted in its briefings to the committee that tests of all three treatment alternatives have demonstrated their ability to meet functional requirements. On that basis, and with the associated changes in the work programs of the three alternatives and their management, the DOE Technical Working Group (TWG)<sup>1</sup> has produced downselection criteria. These were presented to the committee at its first meeting on 20-21 November 2000 (Harmon, 2000a, b), and represent the basis for this report.

## DOE SELECTION CRITERIA AND GOALS

The TWG and its associated committees and consultants employed systematic and relatively transparent approaches for devising quantifiable evaluation criteria. Using information gathered from other DOE sites and other organizations, they began with twenty criteria and reduced them to the final eleven in an effort to eliminate redundancy and criteria unable to discriminate among the alternatives. The final set of criteria (see [Box 1](#)) was approved by the DOE Office of Environmental Management for use in making recommendations on process downselection.

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<sup>1</sup> The TWG has the lead responsibility for developing recommendations on both research and development (R&D) direction and the bases for subsequent recommendations on process selection. This group, using input from a technical advisory team and the Tanks Focus Area (TFA), interacts with a representative of the DOE Office of Environmental Management responsible for the process development and recommendation for downselection. This representative recommends to the DOE Assistant Secretary for Environmental Management the final determination on the downselection outcome.

## BOX 1

## DOE CRITERIA FOR PROCESS SELECTION AT THE SAVANNAH RIVER SITE

1. **Schedule risk**—Risk to the overall project schedule due to high-risk technology issues not being resolved in time to support downselection [to be made in June 2001].
2. **Project cost reduction potential**—Potential that cost savings in the total project cost can be identified (generally due to flow sheet or equipment arrangement changes that would allow facility footprint reductions).
3. **Life-cycle costs through decontamination and decommissioning (D&D)**—Total costs to complete all salt processing (including HLW system costs). The focus is on life-cycle costs, but the separate components' total project cost and operating cost also are examined for key differences.
4. **Technical maturity**—The overall technical maturity of the process flow sheets (including the required strontium and actinide removal steps). EM-50 [DOE Environmental Management Office of Science and Technology] stages of maturity are applied to each unit operation and the results are averaged.
5. **Implementation confidence**—Amount of relevant process experience (large-scale demonstration or deployment) in the DOE complex and industry for the key equipment used for each cesium removal process. This criterion also includes commercial availability of key components and chemicals.
6. **Minimize environmental impacts**—Comparative assessment of environmental impacts from secondary waste streams, airborne emissions, and liquid effluents. This criterion also includes the number of Saltstone vaults required for each process.
7. **Impacts of the interfaces at the Defense Waste Processing Facility (DWPF)**—Cost of implementing the changes (physical modifications) to the interfacing systems and the loss of [glass] canister production caused by outages for equipment installation or transfer line tie-ins.
8. **Process simplicity to interfacing systems**—The simplicity of interfacing the alternative cesium removal processes with other high-level waste systems. The simplicity is measured by the number of process unit operations needed for the interface times a difficulty factor for each interface unit operation.
9. **Levels of safety control mitigation**—Number and type (e.g., passive, active, administrative, preventive, and mitigative) of controls required to maintain the facility in a safe configuration and to protect the worker, public, and environment.
10. **Maximize process flexibility in throughput**—Capability to operate the process at a higher or lower throughput (turn-up or turn-down) based on the equipment in the current pre-conceptual designs.
11. **Maximize process simplicity (operability)**—Simplicity of the process as indicated by the number of pieces of equipment (in both the non-radioactive areas and the remotely operated area) and number of jumpers (piping connections) required inside the remotely operated area.

SOURCE: Harmon, 2000a, 2000b (viewgraph on p. 20 entitled “Criteria Weights—Case A”), and H. Harmon, DOE, email communication, January 5, 2001.

In recognition of some commonalities, the eleven criteria for process selection were grouped by the TWG under the set of six goals shown in [Box 2](#). The criteria were used as a measurement for the effectiveness in reaching these goals.

## BOX 2

### DOE GOALS FOR PROCESS SELECTION AT SRS

**Goal 1: Meet schedule** (Criterion 1)

**Goal 2: Minimize cost** (Criteria 2 and 3)

**Goal 3: Minimize technical risk** (Criteria 4 and 5)

**Goal 4: Minimize environmental safety and health impacts** (Criteria 6 and 9)

**Goal 5: Minimize impact to interfaces** (Criteria 7 and 8)

**Goal 6: Maximize process flexibility** (Criteria 10 and 11)

SOURCE: Harmon, 2000b, viewgraph on p. 14 entitled "Criteria Aligned by Goal"

Other possible goals, such as 'minimize tank space requirements' and 'stakeholder acceptance,' were not included by DOE, because they were considered to be integral to the goals listed above or were not considered to be good discriminators among the alternatives.

The TWG employed a series of steps to develop and implement the proposed criteria. In particular, they used several groups of experts to carry out preliminary application of the criteria to evaluation of the three processing alternatives. This preliminary screening was intended to determine if the criteria were capable of distinguishing among the alternatives and to determine to what extent the outcome might depend on the relative weighting assigned to each of the criteria. In conducting this preliminary screening, each alternative was evaluated by the group of experts and assigned an integer score from 1 (worst score) to 5 (best score). The resulting scores were then normalized to generate 'utility values'<sup>2</sup> that ranged from 0 (worst) to 1 (best). Finally, each utility value was multiplied by a weighting factor ranging from 0.03 (low weight) to 0.14 (high weight); the highest weighting was given to technical risk (Criteria 4 and 5). Finally, a total score for each of the alternatives was calculated by summing the eleven individually weighted utility values.

Several preliminary scoring exercises (carried out by various advisory and management groups of the TWG) were reported at the November committee meeting. In all of the exercises the resulting total scores for the three alternative processes all fell within the range of 0.60 to 0.69; in one exercise the identical total score of 0.63 was calculated for all three alternative processes. The actual scoring and weightings were consensus values arrived at in review meetings among the experts following extensive discussion. This consensus represents the

<sup>2</sup> The utility value is computed by the formula  $u_i = 0.25 (A_i - 1)$ , where  $A_i$  is the score from 1-5 for criterion  $i$ . The total score is then determined by multiplying each utility value by an assigned weighting factor ( $k_i$ ) and summing the weighted scores.  $\text{Total Score} = \sum (u_i k_i)$



informed judgement of these experts. The TWG program plans to reevaluate quarterly the scoring of the alternative processes to take into account the relative progress in the R&D efforts for each alternative. A final downselection decision to one process is scheduled for June 2001.

## COMMENTS ON CRITERIA

Criterion 1: Schedule Risk. The time frame for completion of the cleanup activity could readily be modified by subsequent funding or policy decisions or by environmental issues. Hence, while the criterion is generally useful in broad terms, it may not be a significant discriminator among the processes. It might be preferable to employ this criteria on a 'go/no go' basis, in which it would have zero weight unless the calculated risk exceeded the inherent uncertainty.

Criterion 2: Project Cost Reduction Potential. Cost is an important consideration in any project of this magnitude. The costs assigned to the process are likely to be governed largely by the cost of major new facilities, and DOE has carried out extensive cost estimates. These initial estimates did indicate differences between the three processes, but the uncertainty in these estimates is sufficiently large that the projected costs for the three alternatives may be essentially equivalent.<sup>3</sup> Cost reduction would result from divergence from the estimates, so if these have been carried out consistently (i.e., with the same level of conservatism), it is unlikely that the criterion will discriminate among the alternatives. At this early stage, cost estimates are not very accurate, and from a policy standpoint there may be a difference between capital costs and operating costs that makes the current estimate of life cycle costs inadequate as a factor for decision making.

Criterion 3: Life-Cycle Costs Through Decontamination and Decommissioning. The federal budgeting procedure takes place on an annual basis and does not ordinarily include life-cycle costs. In addition, funding from more than one DOE Environmental Management office complicates the financial aspects of the cleanup. Consequently, while life-cycle cost is an important issue, the high uncertainties in DOE cost estimates may limit its value in decision making unless the project is privatized.

Criterion 4: Technical Maturity. This criterion appears to provide reasonable input for the downselection procedure, since the major uncertainties identified by the previous committee (National Research Council, 2000) were in areas of science and technology.

Criterion 5: Implementation Confidence. This criterion evaluates the extent to which a given technology has been demonstrated or deployed at large scale, with higher scores assigned when previously used for processing radioactive materials or used within the DOE complex. This does not appear to be independent of Criterion 4, and if given too large a weighting, could result in double counting.

Criterion 6: Minimize Environmental Impacts. Any process selected for implementation would need to gain the necessary regulatory approval, which will be a clear "yes/no" decision. While it is an appropriate goal for each of the alternative processes to minimize radioactive and chemical emissions and generation of secondary waste, the process to be selected will either meet regulatory approval or it will not. The minimization of waste streams is closely tied to project

<sup>3</sup> Kenneth Lang, Department of Energy, oral communication, February 22, 2001.

cost, so this criterion may not be independent of Criteria 2 and 3. Compliance with existing regulations is assumed by DOE and the committee, so comparison of environmental impacts beyond regulatory levels does not represent a relevant and useful discriminator among the three processes.

Criterion 7: Impacts of the Interfaces at the Defense Waste Processing Facility (DWPF). The major focus of this criterion is the process interface with the DWPF, and indirectly with the Saltstone Facility, primarily in terms of number of canisters of vitrified waste to be produced. The DWPF probably represents the most complex and schedule-sensitive operation. Technical modification of these interfaces to allow greater system flexibility would seem to be part of Criteria 4 and 5. In addition, the impact of the interface to DWPF will appear in schedule and costs, so this criterion does not appear to be independent of Criteria 1 through 3.

Criterion 8: Process Simplicity to Interfacing Systems. This is similar to the preceding criterion, and the impact of complexity of the interfaces will appear in schedule and costs.

Criterion 9: Levels of Safety Control Mitigation. As in the case of Criterion 6, regulatory approval will be on a “yes/no” basis, and DOE would only select a process that could be operated safely. The impact of any additional levels of safety control mitigation would appear under cost, so this does not appear to provide discrimination among the alternatives.

Criterion 10: Maximize Process Flexibility in Throughput. This criterion is closely related to several others, including Criterion 1 (schedule), Criteria 4 and 5 (technical), Criterion 7 (interfaces), and Criterion 8 (simplicity and interfaces). While the capability to increase throughput above that of the process design may be desirable for cost factors, such enhancement could have a negative impact on the interfaces with the DWPF and Saltstone operations. Hence, the use of this criterion as a discriminator appears to be in isolation of what should be an integrated system of waste processing. This criterion does not appear to discriminate among the alternatives.

Criterion 11: Maximize Process Simplicity (Operability). The role of simplicity in a process is closely coupled to other factors, including schedule (i.e., lower frequency of process upsets), interfaces with other system processes, and technical risk. However, this criterion may be useful in discriminating among extremes in operability and process complexity, especially where certain operations require very high precision in conditions such as temperature or concentrations.

## FINDINGS AND RECOMMENDATIONS

The purpose of this report is to address the first part of the committee's charge: “evaluate the adequacy of the criteria that will be used by DOE to select from among the candidate processes under consideration.” The eleven criteria—and the goals under which DOE has grouped them—are reasonable and appropriate and were developed in a transparent way.

**Finding: The committee finds that DOE's proposed criteria are an acceptable basis for selecting among the candidate processes under consideration; however, as noted**

**in the preceding discussion, some of the criteria do not appear to be independent of others and some criteria appear unlikely to discriminate among the process alternatives.**

The use of the criteria to reach a final decision relies on a methodology that is still evolving. The weighting factors have not yet been decided, and these may need to be adjusted in consideration of the points raised in the previous section about overlap of some criteria or the concepts of go/no go gates and thresholds. In the application of the algorithm to the process alternatives described to the committee there was little discrimination among the alternatives. There was little difference among the total scores, and the ranking appeared to be dependent upon the weighting factors employed. This raises the question of whether the algorithm is capable of providing adequate discrimination among the alternatives. Is it possible that high scores for certain criteria could obscure serious problems in other criteria?

**Recommendation: The committee recommends that the criteria should not be implemented in a way that relies on a single numerical “total score.” Rather than averaging and totaling the scores for each criterion, the various criteria should be seen as relevant to different goals and purposes and should be considered individually. Some of the criteria should be used as “go/no go” gates and some should have thresholds for use.**

Despite limitations in discriminating among the alternatives, the committee recognizes that R&D progress for the several alternative processes may result in changes in the respective scores on the eleven criteria.

**Finding: The committee finds that the current scoring system for individual criteria can be useful for identifying and following the progress of research and development program prior to a final downselection. This could assist in determining where significant further effort is needed for each process.**

The final selection of a process for treating the SRS high-level waste will be a management decision. The final decision rests with the Assistant Secretary for Environmental Management and will be made on the basis of documentation related to the eleven criteria discussed here. The committee believes that the proposed criteria can provide adequate information for making a risk-informed decision evaluating the science, technology, operational aspects, time factors, costs, and policy matters. As indicated in the preceding comments on the criteria, some issues—for, example, life-cycle costs—do not match well with the federal procedure for allocating funds. This would not be the case for a privatized operation, and if a contractor were responsible for costs it might be necessary for them to be involved formally in the decision-making procedure.

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## APPENDIX A

### BIOGRAPHICAL SKETCHES OF COMMITTEE MEMBERS

**MILTON LEVENSON** (*Chair*) is a chemical engineer with over 48 years of experience in nuclear energy and related fields. His technical experience includes work in nuclear safety, fuel cycle, water reactor technology, advanced reactor technology, remote control technology, and sodium reactor technology. His professional experience includes positions at Oak Ridge National Laboratory (research and operations), Argonne National Laboratory, the Electric Power Research Institute (first director of nuclear power), and Bechtel (last position was vice-president of Bechtel International). Mr. Levenson is the past president of the American Nuclear Society and a fellow of the American Nuclear Society and the American Institute of Chemical Engineers. He is the author of over 150 publications and holds three U.S. patents. He was elected to the National Academy of Engineering in 1976. Mr. Levenson has served on many National Research Council committees, and in 1998 served as principal investigator for the Board on Radioactive Waste Management project on aluminum spent fuel.

**GREGORY R. CHOPPIN** (*Vice-Chair*) is the R.O. Lawton Distinguished Professor of Chemistry at Florida State University. His research interests include nuclear chemistry, physical chemistry of actinides and lanthanides, environmental behavior of actinides, chemistry of the f-elements, separation science of the f-elements, and concentrated electrolyte solutions. While at Lawrence Radiation Laboratory, University of California, Berkeley, he participated in the discovery of mendelevium, element 101. Dr. Choppin's research interests have been recognized by the American Chemical Society with its Award in Nuclear Chemistry and the Southern Chemist Award, the Manufacturing Chemists Award in Chemical Education, and a with a Presidential Citation Award by the American Nuclear Society. He has served on numerous NRC committees, is currently a member of the Board on Radioactive Waste Management, and recently completed a 6-year term as a member of the Board on Chemical Sciences and Technology.

**JOHN E. BERCAW** is the Centennial Professor of Chemistry at the California Institute of Technology. Dr. Bercaw is an expert in organometallic chemistry. His research interests include synthetic, structural, and mechanistic organotransition metal chemistry, compounds of early transition metals, and hydrolyzation of alkanes by simple platinum halides in aqueous solutions. Dr. Bercaw is a former chair and executive committee member of the American Chemical Society's Inorganic Chemistry Division. He is a fellow of the American Association for the Advancement of Science and of the American Academy of Arts and Sciences. His work has been recognized by the American Chemical Society with its Award in Pure Chemistry, the George A. Olah Award in Hydrocarbon or Petroleum Chemistry, and the Award for Distinguished Service in the Advancement of Inorganic Chemistry. Dr. Bercaw was elected to the National Academy of Sciences in 1990.

**DARYLE H. BUSCH** is the Roy A. Roberts Distinguished Professor of chemistry at the University of Kansas. His research, which fathered synthetic macrocyclic ligand chemistry and created the molecular template effect, is presently focused on homogeneous catalysis, bioinorganic chemistry, and orderly molecular entanglements. He is a recipient of the American

Chemical Society's Award for Distinguished Service in Inorganic Chemistry and its Award for Research in Inorganic Chemistry. Recently Dr. Busch received the International Izatt-Christensen Award for Research in Macrocyclic Chemistry and the University of Kansas's Louis Byrd Graduate Educator Award. Dr. Busch was elected president of the American Chemical Society in 2000.

**JAMES H. ESPENSON** is Distinguished Professor of Chemistry at Iowa State University and program director of molecular processes at DOE's Ames Laboratory. He has received the John A. Wilkinson award for excellence in teaching, an award from the Alfred P. Sloan Foundation, and is a fellow of the American Association for the Advancement of Science. He has served as a member of the executive committee and as a councilor for the American Chemical Society's Division of Inorganic Chemistry. Espenson studies transition metal complexes as catalysts for chemical reactions (including oxidation-reduction reactions), as participants in atom-transfer mechanisms, as reagents in new reactions, and as templates for coordination phenomena. His research has focused on oxo- and thio-complexes of rhenium in high oxidation states.

**GEORGE E. KELLER II**, since retiring as a senior corporate research fellow from the Union Carbide Corporation in 1997, has been active in economic-development enterprises and consulting. He is also an adjunct professor of chemical engineering at two universities. His technical expertise lies in separation processes, reaction engineering and catalysis, energy use minimization, and new process configurations. Dr. Keller has 35 publications and 21 co-held patents, and has given invited lectures in many universities, technical meetings, and companies around the world. He is the recipient of four national awards for technical excellence: three from the American Institute of Chemical Engineers and the Chemical Pioneer Award from the American Institute of Chemists. He was elected to the National Academy of Engineering in 1988 and presently serves as a member of the Board on Chemical Sciences and Technology of the National Research Council.

**THEODORE A. KOCH** is currently a DuPont fellow (the highest professional title in the company); he is also an adjunct professor of chemical engineering at the University of Delaware. He has spent his entire career developing chemical processes and bringing them from the benchtop to commercial reality. He holds 29 patents and has authored 9 journal articles and 1 book. He is a member of the Catalysis Club of Philadelphia (former program chair and president), the North American Catalysis Society, and the American Institute of Chemical Engineers. Dr. Koch received the Award for Excellence in Catalytic Science and Technology from the Catalysis Club of Philadelphia and the Lavoisier Award for Technical Excellence from the E.I. du Pont de Nemours and Company.

**ALFRED P. SATTELBERGER** is the director of the Chemistry Division at Los Alamos National Laboratory. Dr. Sattelberger's research interests include actinide science, technetium coordination and organometallic chemistry, and metal-metal multiple bonding. Prior to his current position Dr. Sattelberger held a professorship at the University of Michigan. He is a past member of the executive committee of the Inorganic Chemistry Division of the American Chemical Society, and serves on the board of directors for the Inorganic Synthesis Corporation and the Los Alamos National Laboratory Foundation. He served as a reviewer on the FY 1996 general inorganic chemistry Environmental Management Science Program merit review panel

and on the National Research Council's Committee on Building an Effective EM Science Program.

**MARTIN J. STEINDLER'S** last position was as director of the Chemical Technology Division at Argonne National Laboratory. His expertise is in the fields of nuclear fuel cycle and associated chemistry, engineering, and safety, with emphasis on fission products and actinides. He also has experience in the structure and management of research, development, and deployment organizations and activities. Dr. Steindler has been a consultant to the Atomic Energy Commission, the Energy Research and Development Agency, and various Department of Energy laboratories. He chaired both the Materials Review Board for the DOE Office of Civilian Radioactive Waste Management and the U.S. Nuclear Regulatory Commission Advisory Committee on Nuclear Waste. Dr. Steindler has served on several National Research Council committees, and currently serves on the Board on Radioactive Waste Management.

## **APPENDIX B**

### **LETTERS OF REQUEST FOR THIS STUDY**





Department of Energy

Washington, DC 20585

April 13, 2000

Dr. Kevin D. Crowley Director Board on Radioactive Waste Management National Research Council 2001  
Wisconsin Avenue, N.W. Washington, DC 20007 Dear Dr. Crowley:

I would like to take this opportunity to thank you and your Committee Members for your extraordinary effort providing the Department with an independent technical review of alternatives for processing the high-level radioactive waste salt solutions at the Savannah River Site. We agree with your interim comments noting that additional research and development is required for each option, and we are proceeding with addressing your comments in our research and development plans for fiscal years 2000 and 2001. I am looking forward to receiving your final report this month so that we can make adjustments in the current plans if needed.

I believe that the complexity of the salt processing technology alternatives warrants your continued involvement in our continuing research and development efforts. Therefore, I would like to request that you and your Committee continue to support the Department throughout the next year by providing us with your independent review of each technology road map, and the selection criteria.

Dr. Huntoon has tasked me, as the Deputy Assistant Secretary for the Office of Project Completion, to provide the leadership and program management for technology development and selection of a preferred treatment alternative. I am working closely with the Office of Science and Technology, as well as the DOE-Savannah River Operations Office, to make sure that this effort is adequately supported. An Action Plan has been prepared, and is enclosed, which provides details of the roles and responsibilities for the project. I will be providing the Assistant Secretary for Environmental Management with quarterly progress reviews on each of the technology activities throughout the ensuing months, and I propose that we follow those reviews with a briefing to your Committee to keep you abreast of the salt processing project's progress. Of course, additional briefings, meetings, and documentation will be made available to the Committee as you deem necessary to support your review.

Based on the current schedule, we would be seeking your Committee's review of the items identified above in early summer 2000. Due to the short time available between now and the anticipated time we require your support, may I suggest that you utilize your existing Committee to expedite matters.

Mr. Ken Lang of my staff will be contacting you directly to coordinate the details. Mr. Lang can be reached at (301) 903-7453.

Thank you in advance for your continued support of DOE. I look forward to working with you in this endeavor.

Sincerely,

A handwritten signature in black ink, reading "Ralph E. Lightner" in a cursive script. Below the signature, the word "For" is written in a similar cursive style.

Mark W. Frei Deputy Assistant Secretary for Project Completion Office of Project Completion

Enclosure

cc:

M. Gilbertson, EM-52

K. Picha, EM-22

G. Rudy, DOE-SR

K. Gerdes, EM-54

B. Spader, DOE-SR

J. Case, DOE-ID



Department of Energy

Washington, DC 20585

June 15, 2000

Dr. Kevin D. Crowley

Director

Board on Radioactive Waste Management National Research Council 2001 Wisconsin Avenue, NW Washington, D.C. 20007

Dear Dr. Crowley:

Thank you for your May 16, 2000, letter responding to my request that the National Research Council continue its support of the Department's high-level waste salt processing alternatives at the Savannah River Site.

I am pleased that you would like to continue to provide technical assistance to the Department throughout the planned research and development phase of this project, pending approval of the Board on Radioactive Waste Management and the National Research Council Governing Board.

Your proposals to (1) comment on the criteria that will be used to select a processing alternative; (2) evaluate the results of the research and development work that is undertaken on the candidate processing alternatives; and (3) provide the Department with an assessment of whether the technical uncertainties have been sufficiently resolved to proceed with downsizing the list of alternatives will meet our needs throughout the remaining research and development period. I found the interim report you provided on your current evaluation to be particularly useful in planning the research and development now underway, and I am confident that an interim report for this phase of the study will be valuable in the selection of alternative processing technologies.

Mr. Kenneth Lang of my staff is available to support you and the committee for this review. Mr. Lang can be reached at (301) 903-7453.

Thank you for your continued support of DOE.

Sincerely,

A handwritten signature in black ink, appearing to read "Mark W. Frei". The signature is fluid and cursive, with the first name "Mark" being the most prominent.

Mark W. Frei  
Deputy Assistant Secretary for Project Completion  
Office of Environmental Management

cc:

M. Gilbertson, EM-52  
K. Picha, EM-22  
G. Rudy, DOE-SR  
K. Gerdes, EM-54  
G. Boyd, EM-50  
B. Spader, DOE-SR  
J. Case, DOE-ID

## APPENDIX C

### HIGH-LEVEL WASTE AT THE SAVANNAH RIVER SITE

During and immediately following the Second World War, the U.S. Government established large industrial complexes at several sites across the United States to develop, manufacture, and test nuclear weapons. One of these complexes was established in 1950 at the Savannah River Site (SRS) to produce isotopes, mainly plutonium and tritium, for defense purposes. The site is located adjacent to the Savannah River near the Georgia-South Carolina border and the city of Augusta, Georgia, and comprises an area of about 800 square kilometers (~300 square miles).

The SRS was host to an extensive complex of facilities that included fuel and target fabrication plants, nuclear reactors, chemical processing plants, underground storage tanks, and waste processing and immobilization facilities. Plutonium and tritium were produced by irradiating specially prepared metal targets in the nuclear reactors at the site. After irradiation, the targets were transferred to canyon facilities, where they were processed chemically to recover these radionuclides. This processing resulted in the production of large amounts of highly radioactive liquid waste, known as *high-level waste* (HLW), that, after treatment with caustic, is being stored in two underground tank farms at the site.

#### TANK WASTE PROCESSING

DOE has the responsibility for waste management at SRS and has implemented a program to stabilize this HLW and close the tank farms. The tank waste processing system at SRS comprises the major components; (a) waste concentration and storage, (b) radionuclide immobilization, (c) extended sludge processing, (d) salt processing, and (e) salt disposal.

#### Waste Concentration and Storage

The high-level waste resulting from operations in the chemical processing canyons is currently being stored in 48 underground carbon-steel tanks. The tanks range in size from about 3 million to 5 million liters (750,000 to 1.3 million gallons). The HLW was made alkaline with sodium hydroxide (NaOH) and formed a caustic sludge before being transferred to the tanks to reduce corrosion of the carbon steel primary containment. Consequently, the waste has a high pH (>14) and a high salt (especially sodium) content.

Approximately 400 million liters (100 million gallons) of HLW were produced at SRS since operations began in the 1950s, but this volume has been reduced to about 130 million liters (34 million gallons) by removal of excess water through evaporator processing operations. About 10 percent of the waste by volume is in the form of a water-insoluble precipitate, or *sludge*, that contains most of the actinides (i.e., uranium as well as transuranic elements) and strontium-90. This sludge was formed by natural settling and by precipitation when NaOH was added to the waste. The remaining waste consists of solid sodium salts (*saltcake*) and an aqueous solution (saturated with sodium salts) called supernate (which contains approximately 95 percent of the cesium in the tank waste, as well as minor amounts of actinides). The saltcake, produced by crystallization after the alkaline waste was processed through evaporators to reduce the volume of material, will dissolve when additional water is added during waste processing. The saltcake

and sludge contain substantial quantities of supernate within their mass; this interstitial supernate corresponds to about half of the total supernate in the tanks.

### **Radionuclide Immobilization**

The Defense Waste Processing Facility (DWPF) was constructed to immobilize radioactive waste in borosilicate glass for eventual shipment to and disposal in a geological repository. The glass-making process is referred to as *vitrification*. This glass is produced by combining the processed HLW (the processing operations are discussed below) with specially formulated glass frit and melting the mixture at about 1150 °C. The molten glass is then poured into cylindrical stainless steel canisters, allowed to cool, and sealed. The DWPF canisters are about 60 centimeters (2 feet) in diameter and about 300 centimeters (10 feet) in length and contain about 1,800 kilograms (4,000 pounds) of glass. About 700 canisters have been produced to date<sup>4</sup>, and SRS estimates that a total of about 6,000 canisters would be produced by 2026, when the tank waste processing program is planned to be completed. These canisters are to be stored at the site until a permanent geological repository is opened and ready to receive them.

### **Extended Sludge Processing**

Extended sludge processing is being used to prepare the sludge portion of the tank waste for processing into glass. The sludge is removed from the tanks by hydraulic slurring and washed to remove aluminum and soluble salts, both of which can interfere with the glass-making process. The washed sludge is transferred to the DWPF for further processing before being incorporated into glass. Sludge processing would result in immobilization in glass of nearly all of the strontium and actinides from the tanks.

### **Salt Processing**

Salt processing would be used to remove much of the radionuclides from the HLW salt for eventual vitrification. The salt is to be redissolved and transferred out of the tanks. It would then be mixed with a sorbent to remove any remaining actinides (mainly uranium and plutonium) and strontium. The currently planned sorbent is monosodium titanate (MST). The solution will then be subjected to another (and as-yet undetermined) process to remove cesium. The separated actinides, strontium, and cesium would be washed to remove soluble salts and sent to the DWPF for immobilization.

### **Salt Disposal.**

A variety of secondary waste streams are formed during the processing operations described above. Some of these waste streams are recycled back to the tanks, some are recycled within the various processing operations, and yet other wastes are treated and stabilized for burial. Most notably, the “decontaminated” salt supernate (i.e., the solutions remaining after actinide, strontium, and cesium removal) would be disposed of onsite in a waste form known as *Saltstone*. The residual solutions are classified as “incidental waste” from the processing of HLW. Saltstone is created by mixing the residual salt solutions with fly ash, slag, and Portland

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<sup>4</sup>Since this appendix was originally published, over 300 additional canisters have been produced.

cement to create a grout slurry. This slurry is then poured into concrete vaults, where it cures (solidifies) and is eventually covered with soil. The Saltstone contains small quantities of some radionuclides.

### **CESIUM REMOVAL PROBLEM**

As noted above, SRS planned to remove actinides, strontium, and cesium from the salt solutions in two processing steps. First, actinides and strontium were to be removed by mixing the salt solutions with MST, resulting in the sorption of actinides and strontium. The product of this reaction could be removed from the salt solutions by filtration for subsequent processing and immobilization. Subsequently, the removal of cesium from the salt solutions would be accomplished by a yet-to-be-chosen process from among precipitation, ion exchange or solvent extraction processes.

In the late 1970s and the 1980s, SRS developed a process for removing cesium from salt solutions through a precipitation reaction involving sodium tetraphenylborate (NaTPB) and cesium to form cesium TPB (CsTPB): SRS refers to this process as "In-Tank Precipitation." The NaTPB was to be added directly to a large waste tank to produce a cesium-bearing precipitate. SRS undertook an ITP pilot project in 1983 to demonstrate proof of principle. The process removed cesium from the salt solution, but it also resulted in the generation of flammable benzene from radiolytic reactions and possibly from catalytic reactions with trace metals in the waste. In September 1995, SRS initiated ITP processing operations in a tank that contained about 1.7 million liters (450,000 gallons) of salt solutions. The operations were halted after about 3 months because of higher-than-expected rates of benzene generation. SRS staff then initiated a research program to develop a better understanding of the mechanisms of benzene generation and release. They also considered possible design changes to handle the benzene during processing operations and catalyst poisoning strategies.

In 1996, the Defense Nuclear Facility Safety Board (DNFSB) issued Recommendation 96-1, urging DOE to halt all further testing and to begin an investigative effort to understand the mechanisms of benzene formation and release. Investigations by SRS in 1997 uncovered the possible role of metal catalysts in the benzene formation process. SRS concluded, however, that both safety and production requirements could not be met, which led to the suspension of operations altogether in early 1998. At the time of suspension, SRS had spent almost a half billion dollars to develop and implement the ITP process. In March 1998, Westinghouse Savannah River Company (WSRC) formed a systems engineering team to identify alternatives to the ITP process for separating cesium. This team began by undertaking a literature and patent screening procedure to identify currently known processes, followed by a system of analyses by panels of experts to reduce the number of alternative processes to four.

### **Strontium/Actinide Removal by MST**

In all four of the final candidate processes for cesium separation, prior removal of strontium and actinides is viewed by SRS as a requisite process. At present, the use of MST is the method of choice. Some technical uncertainties remain to be resolved, of which the major ones are the kinetics of sorption on MST and the amount of titanate acceptable for proper quality of the vitrified waste form.

### Tetraphenylborate Precipitation Process

The ITP developed by WSRC removes cesium from HLW supernates by precipitation with tetraphenylborate ion,  $[B(C_6H_5)_4]^-$  (TPB). Sodium TPB is a reagent used for analyzing for the potassium ion based on the insolubility of potassium TPB (KTPB). The 200-fold lower solubility of cesium TPB (CsTPB) can provide decontamination factors (DF) from the salt as high as  $10^5$  to  $10^6$  and the mixed CsTPB/KTPB precipitate is typically in a form that is easily filtered. On the average, the SRS HLW in the waste tanks contains sodium ions (approximately 5 molar), potassium ions (approximately 0.03 molar), and cesium ions (approximately 0.00025 molar).

HLW treatment, including the removal of cesium-137, involves separation of selected radioactive components and their subsequent immobilization in a borosilicate glass at the DWPF. To prevent organic material from being fed to the DWPF melters, the CsTPB/KTPB precipitate must be treated to remove more than 90 percent of the phenyl ( $C_6H_5$ ) groups bound to the boron. Thus, a precipitate hydrolysis process (PHP) was developed to hydrolyze the TPB using formic acid in the presence of a copper catalyst. The hydrolysis products are benzene, which is removed by evaporation and incineration, and an aqueous solution containing  $^{137}Cs^+$ ,  $B(OH)_3$ , and  $K^+$  ions. An attractive feature of TPB is its susceptibility to catalytic decomposition downstream.

### Crystalline Silicotitanate Ion Exchange

Ion exchange has been in commercial use for over 100 years to remove ions from aqueous solutions, e.g., to make deionized water. In most applications the separated ions are eluted from the ion exchange material, e.g., using a dilute acid, the eluted ions are concentrated, and the ion exchanger is reused over and over. Although this technology is well established, ion exchange for cesium removal from high-level waste at SRS and other DOE sites poses challenges. The ion exchange material must withstand both high alkalinity and high radiation fields and must be very selective for cesium in the presence of much higher concentrations of the chemically related sodium and potassium ions. A promising material for use by SRS to remove cesium is crystalline silicotitanate (CST), developed by Sandia National Laboratory and Texas A&M University, based on work performed on amorphous hydrous titanium oxide in the 1960s and 1970s at Sandia. CST has received considerable attention because of its promise as an ion exchange material for nuclear waste applications. The material has a high selectivity for  $Cs^+$  in salt solutions over a large portion of the pH range from acidic to basic solution, and exhibits high stability to radiation as well. CST is also unusual in that cesium is difficult to remove from the material (i.e., it is nonelutable and the CST cannot be reused). As a result, CST must be incorporated into the HLW stream along with the radionuclides, and the stability of borosilicate glass with higher concentrations of titanium is an issue that must be addressed.

### Caustic Side Solvent Extraction

A typical solvent extraction process includes four steps. First, a feed stream is contacted with a solvent that is virtually insoluble in the stream. During this contact, one or more components of the stream transfer to the solvent, while other components do not. The loaded solvent, scrubbed to remove minor contaminants and leaving relatively clean solvent plus the



component(s) to be finally recovered, is sent to a stripping operation where the component(s) to be recovered is removed. The stripped solvent may then go to a solvent-recovery step, in which it is cleaned prior to returning to the first step. In such a process, very high removals of extracted components often can be attained.

Solvent extraction has had a long history of successful use in the nuclear industry for such operations as spent fuel reprocessing and plutonium recovery. This history includes long periods of time in which solvents of various organic species have been exposed to high-radiation fields without experiencing catastrophic degradation rates. Solvent extraction operations usually consist of selectively transferring components from an aqueous, acidic stream into the organic stream. A second aqueous stream of somewhat different composition is often used to strip the solvent and concentrate the extract. For the SRS application, the solvent extraction process must remove approximately 99.998 percent of the cesium (a decontamination factor, or DF, of 50,000) from an aqueous, tank-waste feed stream. The raffinate aqueous stream, thus purified of cesium, would be sent to the SRS Saltstone Facility, and the extract, concentrated in cesium by about an order of magnitude is sent to the DWPF.

### **Direct Disposal in Grout**

Direct disposal of the tank waste following removal of strontium and actinides is very similar to the Saltstone process that was to have been used to dispose of the salt solutions from ITP operations as low-level incidental waste. Although it is a rather mature technology and has already been demonstrated at the site for less radioactive salt solutions, the degree of retention of cesium may not satisfy regulatory requirements.

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