SUMMARY OF FISCAL YEAR 2014 ACTIVITIES: DISPOSAL-RELATED INTEGRATED SPENT NUCLEAR FUEL REGULATORY ACTIVITIES—IDENTIFICATION AND ANALYSIS OF KEY REGULATORY AND TECHNICAL ISSUES FOR DISPOSAL OF SPENT NUCLEAR FUEL AND HIGH-LEVEL WASTE

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QUALITY OF DATA, ANALYSES, AND CODE DEVELOPMENT

DATA: No data are contained in this report, which contains only summaries of technical results

ANALYSES AND CODES: No scientific or engineering software was used in the analyses contained in this report.

1 INTRODUCTION

The Center for Nuclear Waste Regulatory Analyses (CNWRA®) continues to provide technical support to the U.S. Nuclear Regulatory Commission (NRC) in the Disposal-Related Integrated Spent Nuclear Fuel Regulatory Activities (ISFR) program. This program enhances regulatory efficiency and effectiveness as the agency responds to an evolving national policy on the management and disposal of spent nuclear fuel (SNF) and other high-level radioactive waste. Program activities are conducted as CNWRA charter contract Task Order 9 (Job Code J5662)—titled Identification and Analysis of Key Regulatory and Technical Issues for Disposal of Spent Nuclear Fuel and High-Level Waste. The objective of this task order is for CNWRA to provide NRC with technical assistance on the identification and analysis of key regulatory and technical issues associated with a variety of alternatives for managing the back end of the fuel cycle, including ultimate disposal. These activities are needed regardless of the direction of national policy, and were designed to support the Agency's mission under a range of policy outcomes—all of which are likely to require geologic disposal as a key component.

During fiscal year 2014, the team's independent technical investigations focused on selected key technical issues associated with ultimate disposal, and collaboration with NRC continued on the development and application of a system-level model of the back end of the fuel cycle. These activities were conducted as six subtasks, each focused on analyses of a particular technical topic and each conducted by a team of CNWRA and NRC staff members. Work on these topics builds on previous years' activities regarding risk-significant features and processes and efforts at familiarization with alternative geologic repository host rocks and designs, with a focus on those topics most generally applicable and potentially useful in terms of maintaining and enriching the NRC and CNWRA staffs' skills, knowledge, and technical contributions supporting NRC's regulatory responsibilities.

The work rate on this task order during the first half of fiscal year 2014 was affected by two major factors. Early in the year, the timing of renewal of the task order and obligation of associated funding was uncertain, such that work was slowed down in Periods 2 and 3 (roughly November and December 2013) relative to the original plan. Then, late in Period 4 (mid-January 2014), CNWRA began work on a task order to support completion of a safety evaluation report on the license application for a proposed geologic repository for high-level radioactive waste at Yucca Mountain, Nevada. As CNWRA staff resources were diverted, at NRC direction, to the high-priority Yucca Mountain work, activities on Task Order 9 were again affected. As a result, project activities and products were rescheduled; for example, no intermediate milestones were transmitted during the first half of the fiscal year. While work was reduced on all tasks, work on Task 1 was affected the least in order to maintain pace with the international **DE**velopment of **CO**upled Models and Their **VAL**idation Against **EX**periments (DECOVALEX) program. During the second half of the fiscal year, staff availability for Task Order 9 improved and the work rate increased.

This annual report, organized by subtask, briefly summarizes activities conducted from October 2013 through September 2014. CNWRA activities conducted under the precursor projects in fiscal year 2012 and in the first 2 months of fiscal year 2013 were documented in Pickett (2012a,b), fiscal year 2013 Task Order 9 activities were summarized in Pickett, et al. (2013a), and a mid-year report for fiscal year 2014 may be found in Pickett, et al. (2014).

2 TECHNICAL ACTIVITIES

In the first half of the fiscal year, the only documented project products were a conference presentation and an abstract (the latter described in Section 2.3). A staff member helped organize a topical session on high-level waste disposal for the annual meeting of the Geological Society of America and presented a talk summarizing project activities (Pickett, et al., 2013b). With the increased activity level in the second half of the year, more products were prepared and are described in the following sections.

2.1 Continued Participation in DECOVALEX

CNWRA continued supporting NRC participation in the international DECOVALEX program by further developing in-house modeling tools and expertise. The major CNWRA modeling task involved coupling the thermohydrological code *xFlo* with the geomechanical code FLAC3D™ to produce the tool to be used in the NRC/CNWRA DECOVALEX task (Task B1). This coupled code will be an important tool for DECOVALEX task activities in future years. CNWRA participated along with NRC staff in the (i) 4th Workshop held in Mont Terri, Switzerland in November 2013 (Manepally and Fedors, 2014) and (ii) 5th workshop held in Avignon, France in April 2014 (Ofoegbu and Fedors, 2014).

DECOVALEX Task B1 is designed to understand coupled thermal-hydrological-mechanical (THM) processes in a bentonite buffer and argillaceous host rock at the Mont Terri underground research laboratory in Switzerland. The task is based on three tests: (i) the HE-D heating test at Mont Terri (host rock only), (ii) a laboratory column test on granular bentonite (used as a buffer for the HE-E test), and (iii) the HE-E heating experiment at Mont Terri (integrating buffer materials and host rock). The activities for this summary mainly focused on developing a numerical model to represent the THM behavior of granular bentonite in a column. The laboratory experiment involves a column that is designed to mimic the boundary conditions for the buffer material in the HE-E test by prescribing temperature of the heater at the bottom and a source of hydration (host rock rewetting) at the top.

The team initially developed a one-dimensional model in *xFlo* and later expanded it to a two-dimensional axisymmetric model to represent the lateral heat loss observed in the experiment. *xFlo* simulations were performed for thermohydrologic processes in bentonite and heat transfer in the experimental apparatus. *xFlo* outputs for the entire simulation period were used as inputs to model the geomechanical processes that focus only on the granular bentonite (one-dimensional column) using the previously developed constitutive model for unsaturated expansive clays implemented in FLAC3D. Discussions during the 5th DECOVALEX workshop indicated that the FLAC3D constitutive model captured the complex relationship between the moisture redistribution and swelling behavior of granular bentonite better than simplistic models used by other participants.

Based on action items identified during a task force meeting at the 5th DECOVALEX workshop, CNWRA is currently focusing on (i) calibrating inputs such as the thermal conductivity, hydraulic conductivity, moisture retention curve, and bulk modulus; (ii) representing shrinkage behavior in FLAC3D; and (iii) increasing the duration of the hydration stage to 15,000 hours for the column test. In addition, the one-dimensional model in the FLAC3D simulation was expanded to two dimensions.

To verify and supplement the *xFlo* model, an alternative model representing only thermal processes was developed using COMSOL. Comparisons of measured temperature and relative humidity at three probe locations with model results indicated that *xFlo*, in general, is able to adequately represent the evolution of moisture redistribution in response to heat and hydration. However, comparison of water intake measurements indicated that the *xFlo* model poorly represented moisture redistribution during the hydration stage, which features large changes in hydraulic conductivity as the granular bentonite swells. Subsequent analysis suggested that accounting for the presence of a porous plate above the bentonite in the *xFlo* model strongly affected the simulation results. Comparison of the measured and calculated axial pressures indicates that the mechanical model adequately represents the mechanical response. Current constitutive relationships, which describe flow in *xFlo*, may need to be revised to account for changes in macro- and micro-porosity of the granular bentonite. Work related to the column test was documented in an intermediate milestone (Stothoff, et al., 2014a), which also discussed current efforts to improve the representation of complex swelling behavior of the buffer in numerical models by processes at a pore scale (macro and micropores). THM modeling capabilities are being enhanced in preparation for the next step of Task B1, which involves modeling the HE-E Test. The HE-E Test focuses on THM behavior of the buffer and its interaction with the host rock. Several published conceptual models and constitutive relationships accounting for flow at all pore-scales were examined in the report. One or more of these models could be used to augment *xFlo* capabilities. The report also included staff considerations regarding (i) several potential augmentations to *xFlo* and (ii) procedures to better link *xFlo* and FLAC. This work was also documented in two abstracts submitted to the 2015 International High-Level Radioactive Waste Management Conference (Manepally, et al., 2014; Stothoff, et al., 2014b).

The continuing development of *xFlo* included resolution of several problems with the code. The team also improved and generalized the unsaturated zone expansive mechanical constitutive model developed using FISH function programming for FLAC3D. Recent work focused on testing *xFlo* to build confidence in simulation results. A suite of 18 test cases was used to compare *xFlo* simulation results with two independent modeling approaches. The two independent approaches matched the *xFlo* results very well for every test problem.

The team is planning to use the MATLAB code as a tool to dynamically link *xFlo* and FLAC3D simulations across the Linux and Windows platforms. (Currently *xFlo* runs only in the Linux environment and FLAC3D runs only in the Windows environment.) An approach for driving *xFlo* across the network (key for iteratively linking *xFlo* and FLAC) was developed and successfully tested.

Efforts will continue to model the HE-E test in preparation for the next DECOVALEX workshop in November 2014. Work related to representing the complex swelling of bentonite accounting for changes at the macro and micro pore scale will continue. Development and verification of the *xFlo* code also will advance.

2.2 Analysis of Waste Form

The objective of Task 2 corrosion studies is to determine SIMFUEL dissolution rates under conditions representative of a predominantly reducing environment in a deep underground repository. The reducing condition includes the presence of dissolved hydrogen and small amounts of dissolved oxidizing species such as oxygen and hydrogen peroxide. The oxidizing species are expected to be present due to alpha radiolysis from decay in the SNF, and hydrogen is expected to be generated from alpha radiolysis and corrosion of the container material. During this fiscal year, CNWRA conducted a literature survey and laboratory experiments. The objective of the literature survey was to identify and summarize prior work regarding the effects of hydrogen on SNF dissolution. Similarly, the laboratory experiments are designed to verify the literature information and conduct an independent assessment of SNF dissolution rates in a reducing environment, and to quantify the effects of varying amounts of dissolved oxygen and hydrogen.

The articles and reports identified in the literature survey—most presenting research results from the Canadian, Swedish, French, and Spanish high-level waste disposal programs—indicate that SNF dissolution rates are significantly lower in anoxic and reducing conditions than in oxidizing conditions. However, the literature varies widely on the dissolution rate under reducing conditions. Some reports suggest that SNF dissolution rates will be completely suppressed in the presence of hydrogen, while others suggest that the effect is more gradual. All studies support a finding that SNF dissolution would be suppressed under reducing conditions compared to oxidizing conditions; however, there is no agreement among the various studies on the extent to which dissolution rates are suppressed.

CNWRA conducted several experiments to determine SIMFUEL dissolution rates as a function of dissolved hydrogen and oxygen levels using an electrochemical cell placed in a glove box. (SIMFUEL is an unirradiated, simulated SNF containing chemically equivalent nonradioactive surrogate elements for fission products, activation products, and actinides.) The studies were conducted in contact with a saline simulated groundwater based on published reference compositions for deep groundwaters in granitic rocks. Experiments involved the following three SIMFUEL types: (i) pure UO_2, (ii) 35 GW-day/MTU burnup equivalent, and (iii) 60 GW-day/MTU burnup equivalent. The solution was purged with various combinations of compressed air and a mixture of 4 percent hydrogen gas plus 96 percent nitrogen gas. For each SIMFUEL specimen, the electrochemical experiments were conducted under the following conditions: (i) saturated oxygen, (ii) saturated oxygen and hydrogen, (iii) unsaturated oxygen and hydrogen, and (iv) reduced oxygen and saturated hydrogen. Electrochemical impedance spectroscopy was used to record the impedance of the working electrode for various combinations of the purging rate of gas and the solution temperature. The collected data were analyzed using an electrical circuit model to estimate the dissolution rates.

One of the key challenges has been achieving particular concentrations of dissolved oxygen for fixed levels of dissolved hydrogen concentration. It was found that the desired oxygen concentration levels can be achieved in the corrosion cell through particular combinations of the H_2/N_2 mixture and compressed air flow rates in the corrosion cell and the N_2 flow rates in the glove box. Another key challenge has been to manage and increase the signal to noise ratio in the electrochemical experiments that measure the SIMFUEL electrode dissolution rates. To this end, CNWRA has identified several parameters that will increase the signal to noise ratio, such as choice of bandwidth, value of the applied perturbation signal to the SIMFUEL electrode, and current measuring resistors. A low-current measurement device was added to the electrochemical current measurement device used for measuring impedance associated with SIMFUEL dissolution.

The experimental work suggests that SNF dissolution rates are dependent on both the dissolved oxidizing species concentration and the hydrogen concentration. The experimental results for the three SIMFUEL specimens show that (i) the dissolution rate increases with increasing burnup for the same condition and (ii) the dissolution rate decreases with decreasing oxygen concentration. The results indicate that while dissolved hydrogen is unlikely to completely suppress SNF dissolution under reducing repository conditions, it will significantly reduce the SNF dissolution rates.

This work was documented in an intermediate milestone (Shukla, et al., 2014) and an abstract submitted to the 2015 International High-Level Radioactive Waste Management Conference (Shukla and Ahn, 2014).

2.3 Analysis of Waste Package

During this fiscal year, an in-depth literature review and related corrosion experiments were completed on copper and carbon steel corrosion under geologic disposal conditions. Tests evaluated (i) hydrogen generation and general corrosion of copper in anaerobic groundwater and (ii) carbon steel corrosion, hydrogen generation, and passivity in anaerobic alkaline water. The carbon steel experiments simulated cement pore water in contact with a carbon steel overpack in a container design that includes a concrete buffer around the waste package. Test durations and temperatures were varied to provide a time history and information on the temperature dependence of corrosion. Duplicate and triplicate specimens were used in all tests to ensure reliability and reproducibility of the results. All the tests were monitored closely for environmental conditions and oxygen concentration level to ensure the high quality of the test results. Preliminary test results were communicated to NRC on a timely basis. For carbon steel exposed to anoxic alkaline water at 30, 50, and 80 °C [86, 122, and 176 °F], corrosion rates derived from the experimental data ranged from 0.1 to 3 μm/yr [0.004 to 0.1 mils/yr]. This is consistent with the range of carbon steel general corrosion rates in an anoxic environment reported in the literature {0.1–10 μm/yr [0.0039–0.39 mils/yr]}. Most of the copper corrosion rates measured by electrochemical methods in O_2-free simulated granitic waters ranged from 1 to 5 μm/yr [0.039 to 0.2 mils/yr]. At this O_2 level (less than 10 ppb dissolved oxygen) the rates are higher than the general corrosion rate ranges of 4×10^{-3} to 2×10^{-2} μm/yr [2×10^{-4} to 8×10^{-4} mils/yr] reported in the literature. Hydrogen was observed to be generated from the corrosion process of both materials. An intermediate milestone was prepared at the end of this fiscal year to summarize the literature review and independent test results for both carbon steel and copper as potential waste package materials (He, et al., 2014a). An abstract and paper summarizing the test results on carbon steel corrosion were prepared, approved by NRC, and submitted for the 2015 NACE corrosion conference (He and Ahn, 2014a,b). An abstract on the most recent tests results on carbon steel and copper experiments was prepared, approved by NRC, and submitted for the 2015 International High-Level Radioactive Waste Management Conference (He, et al., 2014b).

2.4 Analysis of Alternative Geologic Media

Work under this subtask assessed the thermomechanical behavior of salt host rock in a hypothetical underground repository housing heat-generating radioactive waste. In the previous fiscal year, a literature survey of models for simulating the thermomechanical response of intact and crushed salt was conducted (Ghosh and Hsiung, 2013). The survey concluded that a model that accounts for the micromechanical processes affecting salt under loading properly simulates the thermomechanical response of intact salt; further study of the Multimechanism-Deformation (M-D) model was suggested. This survey also concluded that crushed salt initially undergoes consolidation when loaded, with a response similar to intact salt once the relative density of crushed salt approaches that of the intact salt. The Callahan model has been shown to describe this crushed salt consolidation process adequately. It was decided this year to investigate the parameters of both the M-D and Callahan models (29 parameters in all) to assess their relative importance to the performance of the salt host rock in isolating emplaced waste. As a part of this investigation, the closure rate of a room in the Waste Isolation Pilot Plant (WIPP) Facility at Carlsbad, New Mexico will be modeled and results will be compared with actual measurements to improve understanding of parameter significance. Procedures have been developed for estimating the model parameters using results of laboratory creep and consolidation tests. Using these procedures, parameters for both intact and crushed salt at the WIPP facility were determined and compared with the published results. A good match was observed. Subtask work was halted temporarily in January 2014 due to staff involvement with the Yucca Mountain work, and did not resume during fiscal year 2014, but efforts will resume in fiscal year 2015.

2.5 Analysis of Decay Chain Transport

The SOAR code implements a generic model to probabilistically evaluate performance of hypothetical geological disposal concepts for high-level waste. Task 5 involves incorporation of transport of U-238 decay chain nuclides in SOAR. The current official version of SOAR (Version 1.0.03) was updated with four additional daughter products of the U-238 decay chain, namely Th-230, Ra-226, Rn-222, and Pb-210. Version 1.0.03 had been tested during fiscal year 2013 to show the capability of SOAR and GoldSim® (GoldSim Technology Group, LLC, 2013) to properly simulate ingrowth and transport of the U-238 decay chain. Placeholder values were assigned to the solubility, diffusion, and sorption parameters in SOAR Version 1.0.03. During fiscal year 2014, the staff reviewed all the radionuclide transport parameters in the SOAR code, including parameters for the new U-238 decay chain daughter products, and updated their values as part of the effort to develop a new SOAR Version 1.0.04.

SOAR parameters are input to the GoldSim model file through a Microsoft® Access® database. In turn, this database is generated from a set of Microsoft Excel® files, which users modify for data entry. Excel files offer a more intuitive interface than Access files, and Excel files can be well controlled and accessed through a Microsoft SharePoint® system, for efficient collaboration among the NRC and CNWRA staffs. Because of recent version upgrades to Microsoft Access, Excel, and SharePoint, as well as the upgrade of the GoldSim software from Version 10.5 to 11, it was necessary to review the current procedure for linking SOAR to the parameter database. Updates to SOAR Version 1.0.04 are being implemented using GoldSim Version 10.5, because an NRC upgrade to GoldSim Version 11 is pending.

Based on the review of the Access, Excel, and GoldSim version upgrades, the structure of the SOAR parameter Excel spreadsheets was modified and tested to streamline the data entry process and linkage to the SOAR parameter Access database. Two new features of GoldSim potentially useful for future developments of SOAR and other GoldSim-based codes were implemented: (i) the ability to record dates for parameter values and attributes of stochastic distributions in the Access database (thus keeping track of multiple dated entries for any single parameter), (ii) enabling the capability of GoldSim to retrieve and reload specifically dated values from the database using the “Effective Date” feature, and (iii) the ability of GoldSim Version 11 to define distribution functions for input parameters within the linked database (in GoldSim Version 10.5, attributes of distribution functions could only be defined within GoldSim). The new work updating Excel spreadsheets will facilitate implementing these features when NRC upgrades to GoldSim Version 11.

Task 5 products were (i) the SOAR Version 1.0.04 code (not formally transmitted) and (ii) the revised Appendix A (Default Input Parameters) of the SOAR User Guide, including the updated decay chain and other transport parameters (Osidele and McMurry, 2014). The latter intermediate milestone was transmitted in July 2014.

2.6 Fuel Cycle Integration and Coordination

The NRC and CNWRA staffs collaborated on development of a system-level performance assessment model for the back-end of the nuclear fuel cycle (referred to, for brevity, as system-level performance assessment model or SLPA). An objective of this model is to provide a tool to analyze waste management alternatives, enabling staff to examine broad-scale issues that may result from potential changes in national policies on management of SNF. The model provides a methodology to define metrics to quantify system-level impacts related to SNF disposition scenarios. The model is conceptualized as a system of multiple compartments programmed in GoldSim Version 10.5. These compartments represent storage of SNF in (i) pools near nuclear reactors, (ii) onsite dry cask facilities such as independent spent fuel storage installations (ISFSIs), (iii) regional or centralized storage facilities, (iv) reprocessing facilities (i.e., SNF in the queue to be reprocessed), and (v) SNF aging facilities at a hypothetical geologic repository location. The model includes abstractions to estimate:

- SNF masses and their distribution in the five compartments
- Number of canisters in storage facilities
- Average age of SNF in storage facilities
- Mass and cask transfer rates among compartments, as well as the average age of the SNF under transfer
- Dose consequences from routine actions, accidents, external events, and transportation using reference doses gathered from the literature.

The model is a lumped abstraction; for example, storage at reactors is modeled as a single compartment that aggregates the SNF mass at all facilities nationwide. Doses are estimated using existing dose values (e.g., dose conversion factors and dose per metric ton of uranium in SNF) computed elsewhere and scaled by the number of canisters and average SNF activity (which is dependent on the average age of the SNF).

During the first half of fiscal year 2014 the list of SLPA input parameters was consolidated into an annotated Excel file. The objective of this work was to identify areas where information is well defined, areas where the model would benefit from collecting more information, areas where information is not available, and parameters that are hypothetical or related to scenario definition (e.g., future rate of power generation and spent fuel production). A result of this activity was the recognition that steps for loading bare SNF into canisters and casks, as well as the steps related to repackaging (for example to repair degraded casks or canisters), require more detailed definitions in order to quantify the aggregated risk of accidents while performing such steps, or collective worker doses as a result of repackaging activities. The diagram in Figure 2-1 is a visual summary of hypothetical packaging steps.

The team continued to develop and test a repackaging module for the SLPA, programmed as an external, dynamically-linked library (DLL) in FORTRAN. The DLL repackaging module is used for counting the number of casks undergoing repackaging. The repackaging module was updated during this fiscal year to account for the presence of legacy casks and their age. Verifications of DLL module calculations were performed by step-by-step inspection of the results of a simple cask transfer problem, and comparison of results assuming a single age for legacy casks to results assuming a distribution of ages for legacy casks. The module was extended to account for the distribution of ages of the SNF in legacy casks, and output average age of SNF in the five compartments and of the SNF under transfer as functions of time. A

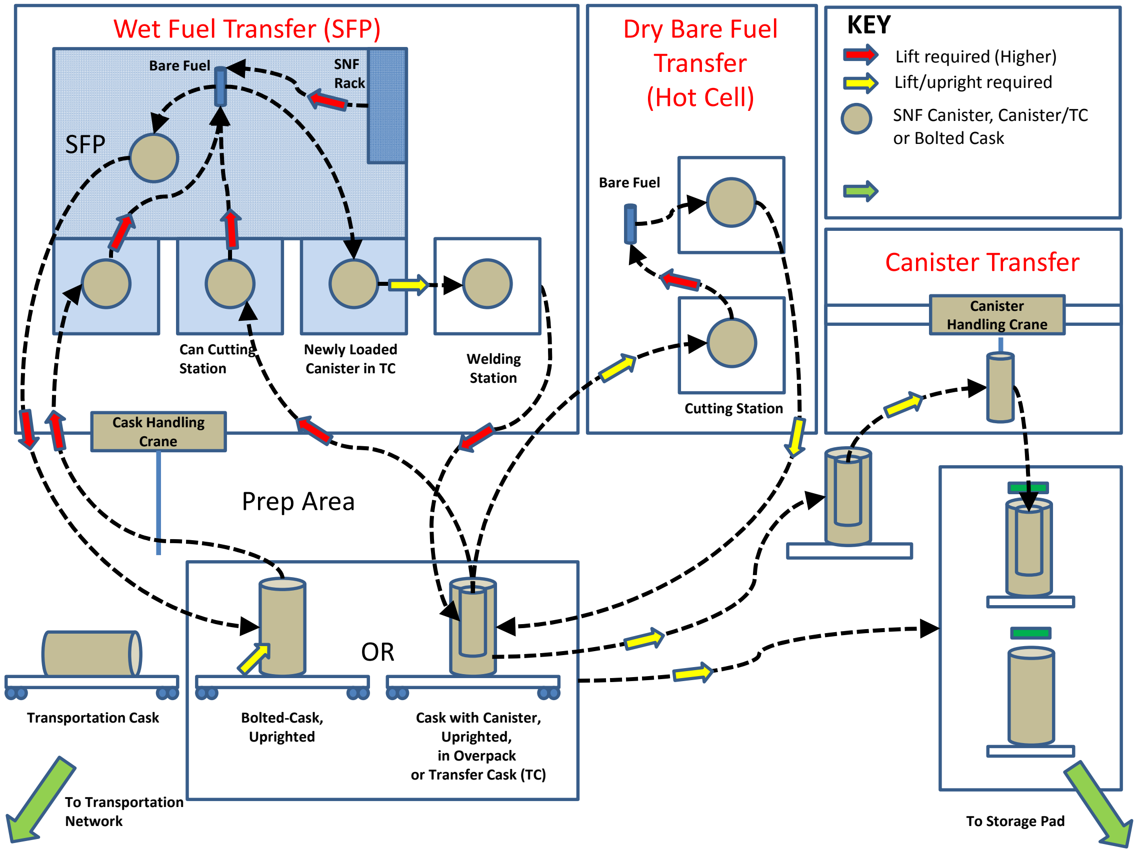

Figure 2-1. Hypothetical Steps for Fuel Loading Into Canisters and Canister Transfer Into Dry Cask Storage Systems

conference abstract describing the repackaging module was prepared, but publication of the material was deferred for programmatic reasons.

Several intermediate versions of the GoldSim model file were released during this fiscal year, starting with Version 56. Version 62 was documented as an intermediate milestone in a transmittal letter that summarized the key updates to the model during the fiscal year (Pickett, 2014). The current model file is Version 64, including the updated repackaging DLL, updated input parameters, and updated approach to quantify doses from accidents and routine activities. A workshop was held September 2014 to describe recent SLPA model updates to NRC staff and discuss basic ideas for future potential applications of the model.

3 SUMMARY AND CONCLUSIONS

Task Order 9 activities during fiscal year 2014, conducted in close collaboration with NRC staff, continued CNWRA contributions to the understanding of risk-significant processes in geologic disposal of SNF and high-level radioactive waste, as well as the potential impacts of alternative conceptualizations of the back end of the nuclear fuel cycle. Technical work was topically organized around coupled process modeling for DECOVALEX, waste form, waste package, salt host rock, decay chain nuclide transport and related SOAR model refinements, and SLPA model updates. Technical work was slowed in response to a shift in resources to the Yucca Mountain safety evaluation report effort, but progress on DECOVALEX and the system-level model was least affected.

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