# HYDROLOGICAL METHODS CAN SEPARATE CESIUM FROM NUCLEAR WASTE SALT CAKE

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#### Abstract

Interstitial Fluid Displacement (IFD) is a new and novel method for separating cesium from saltcake waste. It was identified by reevaluating fundamental factors of the saltcake recovery system, such as physical and chemical properties of saltcake, experience from other technical disciplines and industries, processing rate requirements, process safety requirements, and use of proven-successful processes and equipment.

Chemical theory and limited data indicate that none, or very little, of the cesium is contained in the saltcake crystal structure. Similar logic also applies to potassium and sodium hydroxide, other chemicals important to waste processing. Laboratory and field data demonstrate that saltcake is usually porous, permeable, and dissolves rapidly.

Hydrologic modeling of liquid flow through porous saltcake suggests that the cesium, potassium and sodium hydroxide can be separated at high recovery and low volume using IFD. IFD requires amazingly little equipment—a transfer pump and a few water injection pipes. The benefits include reductions in safety risk and total costs, fewer and simpler unit operations, and improved operability. Cost savings could be significant compared to the baseline process. A holistic or systems approach, where water output from one cleanup operation becomes the water input for another of the waste processing operations, could further reduce the cost of waste tank clean-out.

#### Background

The Savannah River Site (SRS) has 51 tanks containing about 30 million gallons of high level radioactive waste from nuclear fuel reprocessing. This waste must be converted into a stable form for long-term storage. Each tank contains varying amounts of the three waste types: sludge, saltcake and salt solution. The objectives of the waste-processing program are to clean out the waste tanks and produce stabilized waste at the least cost, in the shortest time, and by the safest operation. The operation should also be trouble-free, easy and simple to operate, needing little maintenance and producing negligible secondary

waste. By using tools such as fundamental physics and chemistry, fundamental knowledge of hydrology, water jets and fluidic transfer pumps, a new waste recovery program is proposed with potential to meet the above objectives much better than the baseline process.

In tanks containing predominately saltcake, the radionuclide present in the highest concentration is cesium-137. Cesium's daughter product, barium-137, is a strong emitter of gamma radiation. Pipelines containing solutions of dissolved salt cake typically have high radiation rates. Such pipelines require shielding with several inches of lead or several feet of soil or concrete. Due to (a) the porous nature of the salt cake, (b) the cesium concentration being very much lower than saturation and (c) cesium's ionic radius in nitrate crystals being about 50 percent greater than sodium's; it is quite likely that the cesium-137 exists only in the interstitial liquid. If so, it can be removed from the saltcake by displacing the contaminated interstitial liquid with uncontaminated liquid. Cesium deportment between saltcake and interstitial liquid has not been evaluated previously. However, early studies at SRS of evaporation and crystallization mentioned that cesium seemed to concentrate and to remain preferentially in the salt solution, not reporting to the nitrate salt crystal phase.

If the preferential removal of cesium by IFD is feasible, the saltcake remaining in the tank can be dissolved and pumped through surface pipelines with no shielding, making its removal, processing, and handling much less expensive. It should also be possible to use this low-cesium solution as process water for recovery of sludge making significant water conservation. Experience at the SRS tank farm shows that pipelines transporting waste with a radiation level less than 0.05 Ci/gal generally do not require shielding. In a preceding scoping study, Brooke and Hamm (1) analyzed IFD by simulation. Their simulation showed that hydrological modeling was applicable to saltcake waste tank operation and that IFD would likely work. The simulations reported here are similar but more extensive, more detailed and, it is hoped, more accurate.

#### **Cesium Distribution**

The cesium content varies considerably from tank to tank. Table 1 gives data representative of SRS salt tanks. Most of the cesium mass is distributed in about equal thirds between two tanks at =<25 Ci/gallon, four tanks at =<12 Ci/gallon and six tanks at =<5 Ci/gallon. The cesium solubility in 5.4 molar Na salt solution at 40 °C is 0.715 moles/liter (estimated by Dr. D. McCabe of SRTC using the Environmental Simulation Program by OLI, Inc.). At SRS the Cs-137 is one-third of the total cesium and has an attributed emission of 87 Ci/gram. Thus, a saturated solution would emit 10,738 Ci/gallon. Comparison

with the actual radiation values for SRS salt solutions indicates the Cs levels are only 0.2 to 0.9 percent of the saturation level.

The potassium content is also well below saturation. Potassium solubility in 5.4 molar Na salt solution at 40 °C is 1.60 moles/liter (estimated by Dr. D. McCabe of SRTC using the Environmental Simulation Program by OLI, Inc.). At SRS the potassium in salt supernate is 0.11 to 0.21 moles/liter. Thus, potassium in salt solution is only 7 to 14 percent of the saturation level.

#### Hydrological Modeling

Two types of flow were modeled, drainage and input-output balanced pumping. Drainage is a partially saturated flow without recharging the interstitial fluid. Pumping is a saturated flow with recharge equal to the pump discharge. Pictorial and graphical representations of drainage are given in Figure 1. Drainage proceeds through the four steps shown and ends with a residual saturation, which is the amount of interstitial fluid remaining in the pores after draining. The amount of fluid remaining in the pores increases as the entry pressure increases, which in this case is the gravity head of water. In the residual saturated zone a balance exists between the gravity potential draining the pores and the capillary forces retaining the fluid. In systems like this one where the gravity head is low, the horizontal distances large and the flow rate low; drainage is slow and much of the saltcake will remain saturated for long periods. In an SRS tank only a relatively thin top layer may drain to the lowest possible residual saturation. The relationship between moisture content and pressure head in the saltcake was calculated using the correlations developed by van Genuchten (4) for a typical sand having residual saturation of 17 weight percent fluid.

The flow of water and transport through porous salt cake was modeled using a three-dimensional finite element computer model, FEMWATER (2). The finite element mesh to model the system was created using the Department of Defense (DoD) Groundwater Modeling System (3). Following are the system parameters as modeled:

- An SRS tank with total volume of 1.3 million gallons (4,925 m<sup>3</sup>), saltcake volume of 1.08 million gallons (4,099 m<sup>3</sup>), interstitial porosity or fluid volume of 22 percent (902 m<sup>3</sup>) and saltcake bulk density of 95 lb/ft<sup>3</sup> (1524 kg/m<sup>3</sup>).
- Finite element mesh of 3,021 nodes and 2,030 elements.
- Tank walls represented by no-flow boundary conditions at the walls and bottom of the mesh.
- Drainage simulated by an area of zero pressure head set at seven contiguous nodes on the bottom of the mesh.

- Pumping simulated by assigning to one node the boundary condition of constant flow rate. This node was located 1 m from the bottom of the tank and offset from the wall a lateral distance of 4.5 m.
- An initial condition for all simulations was fluid level at the top of the saltcake representing an initially saturated saltcake.
- Homogeneous permeability equal to a typical sand at 5 x 10<sup>-4</sup> cm/sec (0.018 m/hr) and equal in the x-, y-, and z- directions.
- Heterogeneous permeability represented by two permeability ranges, a wide range of 10<sup>-3</sup>-10<sup>-7</sup> cm/sec and a typical range for sand of 10<sup>-2</sup>-10<sup>-4</sup> cm/sec. Each range was divided into nine steps.

Saltcake permeability represents the greatest uncertainty in the analysis. The magnitude of the permeability controls the flow rate of liquid through the saltcake while the variation in permeability controls the flow pattern. The flow pattern determines how efficiently the interstitial liquid is displaced. Flow will concentrate along paths of high permeability; thus a nonuniform permeability increases the tendency for low flow (dead) zones.

Saltcake forms around the cooling coils more quickly than in the areas between coils leading to a nonuniform salt structure. In addition, hot evaporator bottoms are added periodically to the salt tanks causing dissolution of existing saltcake and crystallization of new saltcake. Photographs of tank interiors show circular saltcake structures around cooling coils, and these structures show noticeable diametrical variation with height. The influence of the cooling coils on the salt structure diminishes with distance and is absent from the material between the coils. Thus, the saltcake is probably quite heterogeneous. Since IFD uses very low flow rates, as long as the overall permeability is adequate for the target flow rate, heterogeneity of permeability is more important than its magnitude.

Integral to this evaluation of IFD is the assumption that, after the final IFD cycle, the remaining saltcake would be dissolved at a 2:1 ratio of water to saltcake. Wiersma (9) determined that this ratio was achieved when adding dissolving water at low flow rates. One measure of success is that the dissolved saltcake solution would be below the pipeline-shielding limit. The other measure of success is that IFD produces a relatively small volume of the high cesium solution (<1,000,000 gallons).

### Drainage Flow

The drainage simulation represents removal of the interstitial fluid by gravity flow with no water added to the tank during draining. The saltcake is drained by pumping from a well that extends through the saltcake to the tank bottom. The pump suction would also be at the bottom of the tank. The interstitial

fluid drains to the pump well by gravity flow. The simulation was run until the pumping (drainage) rate was less than one cubic meter per 100 hours. After draining, the tank was recharged by adding an uncontaminated saturated salt solution at the top of the saltcake. It was assumed that the saltcake media would recharge to 100 percent saturation. Concentration values at each node were calculated after resaturation based on the amount of cesium remaining in the interstitial pore space after the previous drain. At completion of resaturation, the draining process was repeated. Several iterations of the drain-and-recharge simulation were performed. The volume of flow through the system and the amount of cesium removed from the tank were calculated for each cycle. The results are given in Table 2 as simulation numbers 1 and 2. These simulations define a best-case scenario, that is, IFD with uniform flow and drainage patterns. There are only minor differences between the high and low permeabilities. Therefore, only the low permeability was used for succeeding homogeneous condition simulations.

Figure 2 shows the liquid in the saltcake after 300 and 1,000 hours of draining. The bottom layer of saltcake opposite the drain remained saturated during most of the drain process, because the gravity head is inadequate to cause liquid flow to the drain. The volume of liquid drained from the tank was 611 m<sup>3</sup> compared with the original interstitial volume of 902 m<sup>3</sup>. This left 291 m<sup>3</sup> of fluid in the tank or 32 percent of the original fluid volume. The fluid produced was at the initial concentration, as no water was added to the system. This first drain recovered 68 percent of the cesium. The average pumping rate achieved by gravity draining was 2.7 gal/min (0.61 m<sup>3</sup>/hr) based on the volume of fluid removed from the system during 1,000 hr of drain time. After the second drain, a cumulative 83 percent of the cesium was recovered. The third and fourth drains raised the cumulative cesium recovery to 86.6 and 87.2 percent, respectively. The fall-off of in recovery is caused because the liquid follows the same flow pattern as for the first drain. Drain cycles after the third one recover very little cesium.

The dead zone seen in Figure 2 at the bottom of the tank remains at the initial moisture content and initial cesium concentration. The only change in concentration in this zone would be due to diffusion. Dead zones remain fully saturated and at the initial concentration throughout repeating drain cycles with little to no cesium recovery from them. As a result, although the material in the upper portions of the tank may be below the pipeline-shielding limit, the overall concentration in the tank remained above the pipeline-shielding limit.

The next simulations, numbers 3 and 4 in Table 2, evaluated the effect of heterogeneous permeability on drainage. References (5, 6 and 7) deal with the issue of heterogeneity and its effect on dispersive transport. As discussed in those references, permeability is characterized by a random field in which both

the mean and variance of permeability are media properties. The mean controls the flow rate whereas the variance controls the dispersion, which in turn controls the ability to displace the cesium bearing pore fluid. A preliminary simulation was performed on a simple mesh to determine the effects of the heterogeneous salt forming process by assigning lower permeability properties around cooling coil locations. This simulation yielded disconnected low permeability zones in the mesh resulting in numerical difficulties. However, when the permeability was varied randomly through the saltcake matrix, preferential flow paths were established that were very similar to the simulation with the cooling coils while the numerical difficulties were not observed. Thereafter, heterogeneous media was simulated by randomly assigning the permeability values throughout the mesh. The data in Table 2 for simulations 3 and 4 show clearly that heterogeneity has a significant adverse impact on cesium recovery. As the heterogenity becomes more pronounced, the cesium recovery drops and the incremental recovery with additional cycles also drops. This clearly illustrates the direct relationship between heterogenity and development of preferential flow patterns. Figure 3 shows graphically the drainage progress after 1,000 and 3,000 hours. The large dead zone at the bottom of the tank opposite the pump (drain) is quite evident.

The highest level of cesium recovery in the drain simulations is 87.2 percent after four cycles. Dissolving the 4,099 m<sup>3</sup> of saltcake at a 2:1 ratio of water to saltcake would produce 12,297 m<sup>3</sup> of saturated salt solution containing the remaining 12.8 percent of cesium. To remain below the pipeline-shielding limit, the maximum mass of cesium in this solution must be <1.845 kg. Therefore, the maximum initial mass of cesium in the tank must be <14.41 kg, which is equivalent to 5.26 Ci/gal in supernate. Since more than one-half of the SRS tanks contain supernate at about 5 Ci/gal, or less, the IFD process by draining appears promising. A potential benefit is the volume reduction in high level waste compared to the baseline process. The IFD process by draining produces 2,444 m<sup>3</sup> of relatively high cesium content waste. The baseline process would generate 12,297 m<sup>3</sup> of lower cesium content waste, but still requiring high-level waste safety provisions.

Wiersma (9) conducted laboratory tests on draining and pumping interstitial liquid from surrogate saltcake. The IFD simulation reported here was discussed with Wiersma, and he stated, "The results found in the IFD simulation are qualitatively comparable to those determined by my laboratory testing." That is, he observed displacement in his balanced pumping tests, but it was not as effective as the simulation. Unfortunately, his tests used plain water rather than saturated salt solution. Until becoming saturated by the dissolving salt, the added water tends to float on rather than displace the interstitial liquid.

### **Pumping Flow**

Input-output balanced pumping was simulated next and the results are given in Table 2 as simulations 5, 6 and 7. Cesium concentrations in the pump output and the overall tank contamination level were calculated during the simulations. The saltcake was saturated at the beginning of the simulation and it was maintained by keeping a constant liquid level at the saltcake surface, which simulated adding uncontaminated saturated salt solution to the top of the saltcake. Preliminary simulations established the maximum achievable continuous pumping rate at 1.1 gpm (0.25 m<sup>3</sup>/hr). Flow rates higher than this caused negative pressures at the pump, which caused FEMWATER to terminate. Therefore, the flow rate was made 1.1 gpm for all pumping simulations. The results are shown graphically in Figures 4 and 5.

Simulation 5, the homogeneous permeability case (cf. Figure 4), again showed better cesium recovery than simulations 6 and 7, the heterogeneous cases (cf. Figure 5). All three cases showed little additional cesium recovery after the first pore volume displacement. For all three cases the cesium concentration in the pump output decreased markedly as the pumping time increased. The preferential flow paths, which are created by tank geometry and pump placement and exacerbated by heterogeneity in permeability, cause this loss in effectiveness. Unless there is a change in geometry of pump and drain placement or in salt properties, flow of added uncontaminated water persists along the same pathways, from which most of the cesium has been previously removed. In the low flow zones, where diffusion is the principal mechanism for transport, cesium is removed slowly or not at all. Figures 4 and 5, especially 5, show quite large dead zones. Clearly, IFD by pumping flow with surface recharge is not a good design.

#### **Controlled Flood**

The flow patterns developed in the drain and the pumping simulations limited the effectiveness of the IFD process because of the low flow or dead zones. These low flow zones are due to tank geometry and the permeability distribution. Dead zones cannot be avoided with a single pump and surface recharge operation. Multiple pump locations are not practicable at SRS, but multiple recharge wells are. Thus, the possibility of shifting the flow pattern by using one or more recharge wells was simulated. Placing injection wells in the dead zone at the bottom level of the tank to displace high concentration cesium solution is called controlled flood IFD. Controlling the flow patterns by selectively placing injection and production wells to improve flow patterns, and thus recovery, is standard practice in commercial IFD operations such as in situ mining, petroleum secondary recovery and ground water cleanup.

The results of the controlled flood simulations are given in Table 2 as simulation numbers 8, 9 and 10. The results are shown graphically in Figures 6 and 7. Simulation 8 used balanced input-output pumping

with one injection well and one production well. The injection point was at the bottom of the tank, diametrically opposite from the production well. The uncontaminated injection liquid was assumed to have a specific gravity equal to the interstitial liquid, thus eliminating the effects of density on flow. At the beginning of the simulation it was assumed that the saltcake media was fully saturated, represented by the hydrostatic condition with the water level at the top surface of the saltcake media. The saltcake media remained saturated throughout the simulation as equal volumes of liquid were added to and removed from the tank. The overall cesium recovery was much greater in this simulation than in any of the previous drain or pumping simulations due to the location of the injection point.

Simulations 9 and 10 evaluated a combined case of first draining the interstitial fluid and then following with balanced pumping. This simulation was run for both the homogeneous (cf. Figure 6) and the heterogeneous (cf. Figure 7) cases. As expected, cesium recovery was higher in the homogeneous material than in the heterogeneous material. However, the introduction of heterogeneity had little effect on the overall cesium recovery compared with that seen in the drain or pump simulations. The controlled flow option appears quite promising, as the heterogeneous case was able to recover approximately 95 percent of the cesium. These simulations showed that the recharge wells were very efficient at moving water from the dead zones. Figures 6 and 7 clearly show the improved displacement, decreased dead zones and the similarity between the two cases.

Table 3 applies the controlled flood results for the heterogeneous case to the three cesium levels given in Table 1. After removal of 4.7 pore volumes of fluid in the heterogeneous media tanks, 4.7 percent of cesium remains in the tanks. If the saltcake media was then dissolved at a 2:1 ratio of water to saltcake, the tanks with initial concentrations of 5 Ci/gal and 12 Ci/gal would be at contamination levels below the pipeline-shielding limit. The tanks that have an original concentration of 25 Ci/gal are only slightly above the pipeline-shielding limit at the end of the drain/inject/pump operation. The average of all tanks shows that the saltcake recovery could be scheduled to maintain the Cs concentration always below the pipeline-shielding limit.

#### **Confirmation Testing**

The most certain way to determine how IFD will actually perform (per simulation or not?) would be with a test in an actual waste tank. This test would measure the hydrologic parameters of actual saltcake and yield a quantitative assessment of IFD. It would determine exactly how much cesium is recoverable preferentially. It would be not only a test, but also it would be a production demonstration of the controlled flood IFD method. The test would follow these steps:

- After inserting a fluidic pump and collocated fluidic sampler to the bottom of the tank, the supernate would be pumped from the tank until the flow rate becomes low. This measures the permeability and residual moisture of the saltcake. The salt solution produced is predicted to contain >70 percent of the original cesium inventory the tank. The collocated sampler collects composite samples of the liquid pumped from the tank. These samples with the volume data from fluidic pump operation give true mass accounting of the liquid produced.
- 2. The second step of the test-production run would determine how much cesium is recovered with one pore displacement using the controlled flood balanced-pumping operation. This step would determine the level of heterogeneity in the saltcake and the effect of heterogeneity on cesium removal. Again, mass accountability data with respect to time would measure the performance of the IFD process. The simulation predicts that as much as 80% of the total cesium would be recovered by these two steps. The total salt solution produced would be less than one million gallons. This test-production demonstration would establish the effectiveness of IFD for selective recovery of cesium from salt cake.

The hydrological parameters of a surrogate saltcake can also be evaluated in the laboratory at small scale. Unfortunately, the capillary fringe portion of the drainage curve, which is dominated by gravity head, dominates bench-scale tests. Also, development and collapse of dissolution channels cannot be modeled at bench scale because the gravitational stress is proportional to model size. But, if the tests were done in a centrifuge, the gravitational stress would be correctly scaled. The model's dimensions scale as the ratio, N, of centrifugal acceleration to that of gravity. To satisfy similitude for a typical waste tank with 30 ft height using a model having a 1 ft height, a value of N = 30 would be required. The U.S. Army Corps of Engineers has a centrifuge capable of performing this test at its Waterways Experiment Station in Vicksburg, MS. Although gravitational forces can be scaled, there remain issues of the surrogate saltcake's similitude to actual saltcake. These issues include liquid flow rate, dissolution rate and reaction rates. Thus, bench scale testing has significant drawbacks as a verification method.

Mathematical modeling to simulate IFD and salt cake removal should be continued to include several phenomena that are either beyond the current capabilities of numerical models or involve combinations of physical-chemical processes that are not typically found in the same model. The model should be expanded to address the following issues:

- two-phase flow and transport of variable density interstitial water,
- dissolution (deposition) of the solid phase into (from) the water phase,
- heterogeneity of permeability and chemical composition,
- distribution of heterogeneity (domains, tortuosity),

- changes of saltcake properties as dissolution/deposition proceeds, and
- formation of dissolution features such as voids and flow channels.

A mathematical model has two major uses. First, it helps interpret data from in tank or laboratory tests. In particular it will help interpret the demonstration test data. Second, it can evaluate alternate operating schemes, such as the number and location of injection wells for a controlled flood operation. Numerical simulation is a valuable tool for planning operations like this where various scenarios can be explored to determine probable outcomes. The simulator can be applied both to planning operations and communicating plans to field personnel. If field conditions are found to differ significantly from those expected, a simulator helps to diagnose the difficulty and revise the plan as necessary.

#### **Implementation Scenarios**

This numerical analysis suggests that IFD can selectively separate cesium-137 from saltcake. A combination of draining, pumping, and recharging at selected injection locations is the most effective way to remove the cesium-137. This method was the least sensitive to the heterogeneity of the saltcake. By a combination of draining and resaturation of the salt cake, it is believed that (a) the volume of fluid produced can be held quite low, and (b) the remaining saltcake can be recovered as a low level cesium liquid for sludge-removal. This approach minimizes water usage for the operation as a whole. The advantage of this approach is that its success does not depend on some favorable attribute, such as homogeneous saltcake. This operating scheme would have the following attributes:

- The salt cake would be drained initially without recharge. This drainage was estimated to remove approximately 70 percent of the cesium.
- Uncontaminated salt solution would be added through recharge wells to push out cesium stranded in low-flow zones. Using several injection locations, the volume of fluid removed from the system to achieve this goal could be as low as one pore volume. Assuming two to three pore volumes are removed in the cesium recovery steps, the total volume of high cesium solution would be approximately 1 million gallons.
- The salt cake remaining in the tank would produce about 3 million gallons of salt solution with an average cesium concentration below the pipeline-shielding limit.

#### Holistic Operation—Synergism

Process modifications are often evaluated as if there were little to no interactions with other operations in the processing complex, either upstream or downstream. However, because one plant produces feed for

another and plants recycle streams to each other, interactions are expectable and unavoidable. Interactions mean that modifications can produce new and additional effects beyond what was predicted for the original single process. Indeed, the major reason to consider a modification or improvement may not be realized without considering the synergistic or multiplicative effect on the rest of the processing complex. The effects could be negative as well as positive!

Several benefits become available from using IFD for selective cesium recovery that might not have been expected. For example, the high-cesium solution can be used to assure maximum radionuclide levels in the downstream processes. Because of the high-cesium blending solution, evaporator bottoms can be added directly to the pretreatment feed preparation tank, which then eliminates production of new salt cake. Third, the solution made from the cleaned salt cake would be less than 0.05 curies/gallon. At this low radiation level, it should be practicable to distribute it in surface piping to use as the liquid for sludge recovery. This would eliminate use of new process water with its attendant costs of well operation, chemical additions, evaporation, and final disposal. Fourth, because there is 10 times the volume of salt solution as sludge, sludge recovery could be done as a very dilute slurry, under which conditions there should be little risk of pipeline plugging. Fifth, with the reduction in fresh water usage, the evaporator load would be reduced to the point that possibly one of the tank farm evaporators could be shut down. Sixth, the reduced water load would translate to a reduction in saltstone production.

The following criticality concern illustrates improved intrinsic safety from use of IFD. The saltcake can contain insoluble radioactive solids in sufficient quantity that their aggregation could lead to a criticality. In the IFD process, the flow velocity is so low that transportation of the insoluble solids would not occur. The insoluble solids will travel vertically only and, therefore, maintain their areal distribution in the tank. Any of the solids that should fall into the production well would be pumped out continuously by the action of the fluidic pump. The cyclical pump and drain-back action of the fluidic pump would maintain the solids at the bottom of the production well in suspension so they become part of each pump stroke. The water jets used to distribute dissolution water on the saltcake would keep the saltcake collapsing into voids and channels. This helps eliminate preferential flow paths keeping flow velocities low and also eliminating sites for potential accumulation of transported solids.

#### Summary

The application of hydrological science and engineering could change completely the methods used to clean out DOE's waste tanks. The benefits include improved inherent safety, lowered costs, and simpler and easier operations. Hydrological engineering and science have not been applied previously to the

methods used to clean out waste tanks. The work reported here explored the feasibility of IFD through computer simulation of the process. This work built on the conceptual study by Brooke and Hamm (1). The simulations showed that the IFD process could be successful for preferential cesium removal and would produce a salt solution from the remaining salt cake that would be below the pipeline-shielding limit. The IFD process would use a cyclical sequence of draining and resaturation of the salt cake. The cyclical procedure overcomes the effects of heterogeneity on the flow and transport of interstitial fluid. The study recommends a test-production demonstration in an actual waste tank to determine the key parameters of the process and to assess the viability of IFD.

IFD has four desirable characteristics: (a) The method is predictable, simple, easily controlled and requires only a small amount of energy and water to selectively recover cesium. (b) The process uses simple draining with water recharge into low flow zones and a transfer pump to remove the high cesium salt solution. (c) The in tank equipment will require little to no maintenance. (d) IFD has an extensive record of commercial successes in petroleum secondary recovery, ground water clean up and uranium in situ mining.

The IFD process would take place in the following steps:

- 1. The saltcake would be drained initially without recharge. This drainage was estimated to remove >70 percent of the cesium.
- 2. Uncontaminated salt solution would be added through recharge wells to push out cesium stranded in low-flow zones. This step would require the extraction of approximately four pore volumes of highlevel waste if only one recharge well is used. However, by using more than one recharge well, the volume of fluid required could be reduced to as low as one pore volume.
- 3. Assuming four pore volumes are removed in steps 1 and 2, the total volume of high-cesium solution would be <1 million gallons. Steps 1 and 2 would reduce the cesium in the saltcake to a level such that, when dissolved at a 2:1 ratio of water to saltcake, the resulting solution would be below the pipeline-shielding limit.</p>
- 4. The remaining saltcake would be dissolved by spraying water on its top surface using water jets. The water jets would be aimed to collapse saltcake into holes and channels. This will interrupt development of preferential flow paths and clean the tank interior as saltcake removal progresses.
- 5. A fluidic sampler should be collocated with the production pump to collect composite samples of the liquid produced. These samples combined with the volumetric data provided from the fluidic pump operation will give a mass accounting of the materials produced from each waste tank.

The benefits and potential savings that may be realized using hydrological methods to clean salt tanks include:

- <1/2 the cost and time per tank to begin removing waste compared to the baseline process
- no long-shaft pumps, structural steel and support services
- no pump height changes, maintenance, decontamination or disposal
- in tank equipment (water jets, fluidic transfer pumps and fluidic samplers) requires little to no maintenance and can be easily moved tank to tank for reuse
- no initial tank characterization sampling required
- true mass accounting of transferred slurry and liquid using fluidic transfer pump and collocated sampler
- minimum water use
- minimum NaOH use
- produces a low radiation level salt solution for sludge removal
- produces a high specific gravity and viscosity liquid ideal for sludge removal
- cleans tank as waste is removed
- negligible heel remaining
- no well-water system needed; uses discharge water from Effluent Treatment Facility as process water
- simplifies waste recovery operations in the tank farms
- eliminates creation of new salt cake; evaporator bottoms go directly to pretreatment
- significantly reduced evaporator loads; potential to shut down 1 evaporator

At Hanford, IFD removal of the soluble radionuclides could change the issue of tank leakage such that resaturation of the salt cake is allowable for the small volume and time needed to recover the cesium. Following cesium removal, the bulk salt could be dissolved without completely resaturating the salt cake. Modeling would show the hydrological flows produced by different methods of wetting the salt and pumping out the salt solution.

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# **TABLE 1**

#### **Cesium Levels in SRS Salt Tanks**

# of tanks	Supernate (Ci/gal)	Mass of Cs-137 (kg)
2	25	153
4	12	162
6	5	136

# TABLE 2

### **IFD Simulation Results**

SIMU- LATION	PERMEABILITY			CUMULATIVE % CESIUM RECOVERED				
#			flow		$1^{st}$	$2^{nd}$	3 <sup>rd</sup>	4 <sup>th</sup>
	type	permeability	rate	Initial	displace-	displace-	displace-	displace-
		(cm/sec)	(gpm)	drain	ment	ment	ment	ment
Drain simulations								
1	Homogeneous	5 x 10 <sup>-4</sup>	2.7	68	83	86.6	87.2	
2	Homogeneous	1 x 10 <sup>-2</sup>	2.7	71	80	82.0	83.0	
3	Heterogeneous	10 <sup>-3</sup> -10 <sup>-7</sup>	2.7	48	61	64.0	65.0	
4	Heterogeneous	10 <sup>-2</sup> -10 <sup>-4</sup>	2.7	71	82	82.0	82.0	
Pumping simulations								
5	Homogeneous	5 x 10 <sup>-4</sup>	1.1		51	62	69	74
6	Heterogeneous	10 <sup>-3</sup> -10 <sup>-7</sup>	1.1		32	37	40	43
7	Heterogeneous	10 <sup>-2</sup> -10 <sup>-4</sup>	1.1		33	41	46	49
Controlled Flood simulations								
8	Homogeneous	5 x 10 <sup>-4</sup>	1.1		71	80	90	96
9	Homogeneous	5 x 10 <sup>-4</sup>	1.1	68	83	88	93.9	96.5
10	Heterogeneous	10 <sup>-2</sup> -10 <sup>-4</sup>	1.1	71	80	87	92	95.3
Cumulative Volume of Fluid Produced (gallons)								
Drain: simulations 1, 2, 3 and 4			611	1222	1833	2444		
Pump : simulations 5, 6, 7 and 8				902	1804	2706	3608	
Drain and pump: simulations 9 and 10				611	1513	2415	3317	4219

# Table 3

Cesium distributions in SRS salt tanks

Initial Cs Conc (Ci/gal)	Initial Cs Mass (kg)	Cs not recovered by IFD (kg)	Cs Conc after 2:1 Dissolution (Ci/gal)
25	68	3.2	0.085
12	32.8	1.5	0.040
5	13.7	0.64	0.017
Averag	0.036		

#### following controlled flood IFD



FIGURE 1. Pictorial and graphical representation of pore drainage



FIGURE 2. Moisture content of homogeneous saltcake after draining



 $FIGURE \ 3. \ Moisture \ content \ of \ heterogeneous \ saltcake \ after \ draining$ 



FIGURE 4. Cesium content in homogeneous saltcake after balanced pumping



FIGURE 5. Cesium content in heterogeneous saltcake after balanced pumping



FIGURE 6. Cesium content in homogeneous saltcake after controlled flood



FIGURE 7. Cesium content in heterogeneous saltcake after controlled flood