Cryograb: A Novel Approach to the Retrieval of Waste from Underground Storage Tanks – 13501

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ABSTRACT

The UK's National Nuclear Laboratory (NNL) is investigating the use of cryogenic technology for the recovery of nuclear waste. Cryograb, freezing the waste on a 'cryo-head' and then retrieves it as a single mass which can then be treated or stabilized as necessary. The technology has a number of benefits over other retrieval approaches in that it minimizes sludge disturbance thereby reducing effluent arising and it can be used to dewater, and thereby reduce the volume of waste. The technology has been successfully deployed for a variety of nuclear and non-nuclear waste recovery operations.

The application of Cryograb for the recovery of waste from US underground storage tanks is being explored through a USDOE International Technology Transfer and Demonstration programme. A sample deployment being considered involves the recovery of residual mounds of sludge material from waste storage tanks at Savannah River. Operational constraints and success criteria were agreed prior to the completion of a process down selection exercise which specified the preferred configuration of the cryohead and supporting plant. Subsequent process modeling identified retrieval rates and temperature gradients through the waste and tank infrastructure. The work, which has been delivered in partnership with USDOE, SRNL, NuVision Engineering and Frigeo AB has demonstrated the technical feasibility of the approach (to TRL 2) and has resulted in the allocation of additional funding from DOE to take the programme to bench and cold pilot-scale trials.

INTRODUCTION

Cryograb is a technology for the recovery of waste by the freezing of liquid associated with the waste . The frozen liquid forms part of the 'grab' and is therefore a flexible, versatile and efficient method for recovering solids that might otherwise prove difficult to recover.

Freezing of the liquid is achieved by cooling a surface which is placed in contact with the waste, or liquor surrounding the waste. As water transforms to ice, a continuous barrier for contaminant confinement and immobilization is formed. The confinement results in minimal spreading of sediment/sludge during handling and transport (Figure 1) which can otherwise make retrieval of small amounts of solids difficult.

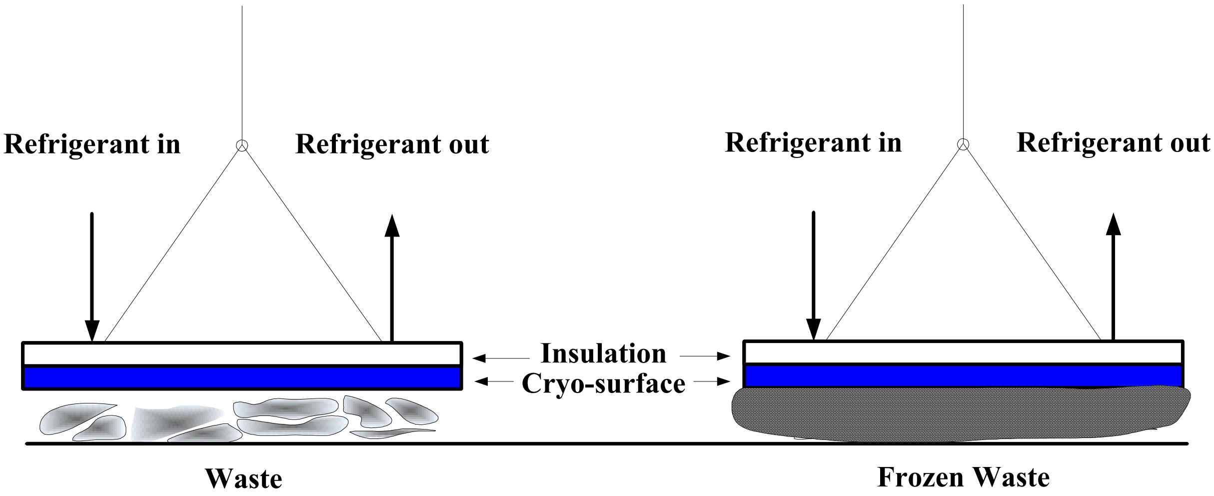

Fig. 1 Cryograb basic schematic

A variety of 'cryohead' grab designs exist which are tailored to the specific requirements of the deployment. These range from 'plates' for large scale recovery of waste, to 'fingers', or collections of fingers for sampling/recovery from difficult locations or even direct injection of refrigerant into waste. A range of options for cooling of the grab surface exist and are selected based on the operational requirements and deployment limitations. Refrigerant options include liquids, gases and electrical cooling.

Once the waste has frozen, the grab is moved with the frozen waste attached to the required destination. The waste is detached by ceasing cooling and awaiting thaw. The thaw may be accelerated by heating the grab.

Cryograb technology has already been deployed successfully in nuclear and non-nuclear applications. Example deployments include the recovery of fragments from within a nuclear reprocessing plant (Figure 2a), the recovery of sludge from a fuel storage pond (Figure 2b) and the recovery of sediment from the sea bed (Figure 2c).

Fig. 2 Cryograb deployment examples

BACKGROUND

The Savannah River Site has 51 underground tanks that are used for storing 138,000 cubic meters (36.4 million gallons) of hazardous and radioactive waste (Figure 3). Four different designs have been used and are called Type-I to Type-IV tanks. All but the Type IV tanks contain dense networks of vertical and horizontal cooling pipes, referred to as cooling coils, which can circulate cooling water. Access to the interior of the tanks is attained through access portals, called risers, which rise from the top of the tank through the ground cover to the land surface. The waste tank bottom is 45 to 50 feet below ground level. The majority of the risers are less than 2 feet in diameter. The area above a tank is typically congested with supporting infrastructure.

Fig. 3 Typical storage tanks at Savannah River Site

The tanks contain waste resulting from reprocessing operations. The types of nuclear reprocessing carried out at SRS include plutonium uranium extraction (PUREX - designed to generate Pu product), H-Modified (HM – designed to generate enriched uranium), and thorium extraction (THOREX). The waste products from these processes differ slightly in composition, but each is a result of a liquid-liquid organic extraction process with ion exchange. PUREX removes plutonium and uranium isotopes from irradiated fuel rods (dissolved in concentrated nitric acid). Similar to PUREX, HM is modified to process limited amounts of other isotopes such as neptunium and californium. Specialized campaigns to recover americium and curium were also performed. THOREX is a method to remove thorium isotopes. Each process carried out in the SRS separations facilities or canyons, is acidic. Because the waste tanks are made of carbon steel, the acidic reprocessing waste stream is neutralized with sodium hydroxide and corrosion inhibited with sodium nitrite in the separation facilities before being sent to the tank farms. The neutralization reaction creates salts and precipitates solids [1].

The composition of the sludge solids depends primarily on the original separations process. For example, the solid waste products from the PUREX process are predominately iron-based compounds with small amounts of uranium and trace amounts of plutonium. The PUREX sludge is characteristically dark brown with quick-settling solid particles. The HM process conversely produces less dense sludge that is aluminum-based, with slight iron, with a longer settling time. Settled HM sludge is difficult to re-suspend because of the cohesiveness of the small particles.

Tank Closure Process

Waste removal from the underground tanks follows a multi-stage process which includes:

- Bulk Waste Removal - extracts the majority of the tank waste, which reduces the radiological and environmental risk inherent in storing large volumes of waste material providing time for subsequent phases to be completed.

- Mechanical Heel Removal - Heel removal is normally accomplished in two steps. The mechanical portion involves non-chemical means such as mixing, spray washing and lancing. The mechanical methods help reduce the amount of chemicals used for the next step. Typically, the mechanical heel removal step continues until it is no longer effective.

- Chemical Heel Removal - Chemical cleaning is the second part of heel removal and assists in removing waste constituents that could not be removed using mechanical methods.

- Cooling Coil Flushing - Cooling coil flushing is designed to remove internal contamination that may have leaked into the coils after years of service. Some of the coils are broken; therefore, any water added to the waste tank from flushing would be removed before the grouting phase. Usually, flushing cooling coils is performed at the same time as heel removal because any flush water is routed to the primary waste tank.

- Annulus Cleaning - Many of the older style waste tanks (Type I and II) have had waste solutions leak from the primary vessel into the annulus. Annulus cleaning for removal of contaminants involves rinsing the outside walls of the waste tank and the annulus pan. For the few waste tanks where more than a simple rinse is needed, mechanical means (e.g., mixing pumps or jets) may be used for the removal of residual waste. Annulus cleaning is done along with the primary tank heel removal activities (including chemical cleaning) because the rinse solution is sent to the primary waste tank, or another waste tank.

- Isolation/Final Sampling - When a waste tank is cleaned and ready to remove from service, waste residue samples are taken from the primary tank and annulus, if applicable, to assist characterization of the remaining radiological and hazardous components.

- Grout Tank - Filling the waste tank with grout ensures final removal from service. Cement-based grout: 1) fills void spaces, 2) reduces the risk of subsidence, 3) provides a physical barrier from weather exposure, 4) is a physical deterrent to casual intruders, and 5) creates a condition that discourages transport of remaining residue.

Significant progress has been made to date with 2 of 12 Type I tanks being emptied, 2 of 4 Type II tanks emptied and 2 of 8 Type IV tanks closed (grout-filled) and 2 tanks emptied in preparation for closure (Type III tanks, 27 still in use). Waste has been recovered by mechanical agitation and pumping. The recovered sludge is vitrified. Where residual solids have remained an additional process of oxalic acid based chemical cleaning has been developed and in combination with hydraulic recovery has proved very effective in removing material from tanks. Due to the complex geometry of certain tanks dead-spots have been encountered where hydraulic recovery becomes challenging and time-consuming. The dead spots result in ‘mounds’ of material ranging from 1-2 inches deep and wide e.g. [2] but in some instances accumulations are up to 1-2 feet deep and wide (Figure 4). It is necessary to remove such material in order to comply with agreed standards for tank closure.

Cryograb has therefore been assessed to support the chemical cleaning phase of the tank closure..

Fig. 4 Example mound of residual material

Waste characteristics

The waste in each tank varies with the history of reprocessing operations and subsequent waste retrieval activities. As a result an illustrative range of properties were agreed for preliminary assessments. These are summarized in Table I below.

Table I. Summary of waste properties.

Property	**Value**	**Comment**
Composition	primary crystalline phases are: hematite, gibbsite, boehmite	Mixtures of Fe, Al oxides and hydroxides, with small amounts of moderately soluble salts (carbonates, oxalates)
Particle size	mean, 0.1-250 µm range, >95% <50 µm	Indicative range agreed for assessment
Density	1-1.9 g/ml	Indicative range agreed for assessment
Water content	5-25 wt. % sludge for typical wet slurries, water content varies depending on liquor composition	Water content values as low as 5% were recently measured for a sample of dried salt removed from the annulus of one tank.
Latent heat	1 - 2 BTU/hr/gal	Typical representative ranges of latent heat for the tanks are 1 - 2 BTU/hr/gal. Latent heat is lower for the F area tank heels. For smaller heel mounds, much heat transfer occurs to the tank walls
Thermal conductivity	3.9 W/mK	Sludge thermal conductivity estimate used for previous sludge drying assessment
Tank temperature	25-50ºC	If pumps were turned off for several days post bulk sludge recovery the temperature would be expected to be near 25ºC
Yield stress	5-45 Pa	Indicative range agreed for assessment
Viscosity	19-35 cP	Indicative range agreed for assessment

DEPLOYMENT SCENARIO UNDER CONSIDERATION

Deployment of cryograb for the recovery of residual mounds of material from Savannah River

Waste Tanks, containing network of cooling coils was selected for initial feasibility assessment. The following assumptions were agreed for use in the feasibility assessment [3].

Operational constraints

Physical constraints

1. The working dimension of accessible risers is 8 inches. This limits the upper dimension for the cryohead and associated equipment that can be introduced into the tanks. A maximum of two adjacent risers can be accessed simultaneously.

2. In order to access the waste mounds the deployment system should have a vertical reach of up to 35 feet and horizontal reach of 12 feet through a complex geometry of cooling coils.

3. The vessel wall temperature should be kept above 21ºC. This relates to the risk of compromising vessel integrity due to possible brittle/ductile transitions that occur below this temperature. The 21ºC limit applies to tanks 1-16 and 29-34. A lower limit of 15ºC applies for tanks 25-28 and sub-zero limits for tanks 38-51.

4. Minor changes in the waste properties would be acceptable (e.g. if the nominal average particle size remained ≤70μm). If significant changes in the physical properties occurred that could impact on slurry transfer through existing equipment or sampling equipment, a formal study including detailed testing might be required unless the volume of material is sufficiently small relative to the bulk material remaining to be processed.

Chemical constraints:

1. Any additives (intentional or unintentional) will require formal review and approval.

2. The waste heels are aqueous radioactive slurries, hence hydrogen will be generated by radiolysis. Since the lower flammability limit (LFL) is 4 volume percent for hydrogen, flammability is always a concern when considering new operations within the waste tanks. It is expected that a compliant system can be engineered and will be based on use of existing approved equipment where possible.

Success criteria

The following success criteria were agreed:

Success criteria 1: Removal of 95% residual material removal in 3 months. The residual material is defined as mounds of material 1-2 feet deep and wide.

Success criteria 2: Recovery of material to within 1-2 inches (~2.5 to 5cm) of the vessel base.

Success criteria 3: Recovery of the waste between or attached to the cooling coils.

OPTIONEERING ASSESSMENT

Cryohead designs and technology to deploy cryoheads were assessed independently to distinguish between the functional requirements of the waste recovery device and the means of deployment.

The cryohead designs considered for this deployment were: flat plate shaped (Figure 5 a); flat plate shaped with additional pokers/prongs protruding from the cryoface (Figure 5 b); articulated mesh (Figure 5c); single and multiple pokers (Figure 5d); cylinder (cup shaped) (Figure 5e); crawler (tank track) (Figure 5f); and floating (articulated plate with a ballast).

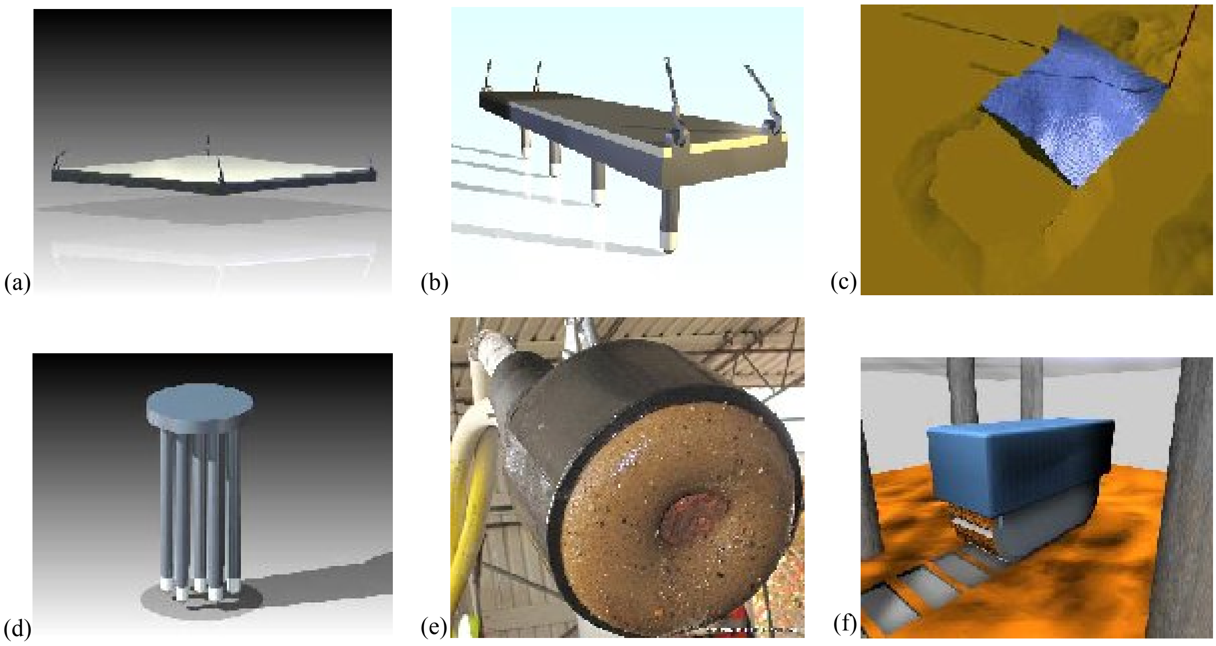

Fig. 5

The deployment design options considered were: conventional rigging; guide frames, ropes, pulleys; simple material handling masts (3 Degrees of Freedom, DoF); complex material handling masts (>3 DoF); kinematically redundant manipulators (>6 DoF); hyper redundant manipulators -snake arms (>6 DoF); mobile vehicle (tracked) – umbilical; mobile vehicle (tracked) - un-tethered; mobile vehicle (floating or submersible) – umbilical; mobile vehicle (floating or submersible) - un-tethered; climbing (limited to wall climbing no cooling coils); and semi autonomous.

Following a review of the options against the defined operational constraints and success criteria (such as the ability to penetrate waste and ability to retain frozen waste upon movement) the preferred system (described below) was defined for further assessment.

PROPOSED SYSTEM DESCRIPTION

The aim of the cryograb system is to coral waste into a single location within the vessel where it can be recovered. The preferred deployment system consists of a vertical mast capable of extending to the bottom of the tank. The bottom section of the mast would be made up of a kinematically redundant or hyper-redundant bespoke manipulator system, capable of maneuvering the cryohead in between the cooling coils on a 12 foot radius from the vertical axis of the riser. The end effector would be a simple gripper with rotation and wrist articulation compatible with a lifting attachment on the cryohead. Closed Circuit Television and lighting would be mounted adjacent to the end effector.

The volume of frozen sludge formed in a cycle has been thermally modeled (see Underpinning Modelling section) and, for a practical cycle time (assumed 12 hours), appears limited by the thermal conductivity of the sludge. Based on expected waste properties and freeze times the total volumes of frozen sludge formed are currently assumed to be limited to 4 inches (~0.1m) from the cooled surface. The preferred configuration is therefore either multiple pokers or a cylinder (Figure 5d and 5e).

For the multiple poker configuration, the head plate is 8 inches diameter (in order to fit through the existing access) and the pokers ½ inch diameter and 12 inches long. The array of pokers presents the opportunity to freeze laterally within the sludge bed (between the pokers) and contrasts with simple plate designs which freeze out parallel from the cryograb. The length of the pokers and spacing can be used to define the mass of material recovered per cycle. The pins are connected to a manifold incorporated into the head plate. The assembly is lowered into the tank and positioned into the sludge.

For an 8 inch restriction the pins are shown on a 4 inch pitch. This arrangement may appear sub-optimal. However, freezing will extend beyond the diameter of the head plate generating a frozen zone of the order of 16 inches diameter. The area, and hence volume of sludge frozen is nominally doubled. The pins may incorporate thermal stand-off tips to minimize temperature reduction at tank walls and thermocouples to monitor wall temperatures or track freezing progress if required. Additionally, it is considered that, at this pitch the assembly can be lowered near or over coils and fittings.

In assessing the coolant requirements of the various cryoheads it is assumed that all are cooled by a refrigerant, glycol/water mixture, supplied by umbilical. From the initial thermal modelling and based on cooled surface area, an umbilical of ½ inch ID provides reasonable flow velocities. At this size the supply and return umbilical would be manageable by the deployment system. Additionally, the total refrigerant volume in the system and hence total loss in event of leak is considered low.

To manage the risk of contaminated refrigerant fluid on the inactive side, the freeze plant and distribution will be divided in two circuits; the primary circuit which connects the freeze plant to heat exchangers and the secondary circuit providing refrigerant to the cryohead. If heating is required to reduce the thaw time a 'hot' glycol/ water mixture can be circulated through the same umbilical. In this way, purging of the system is not required. The thermal modelling also indicates a peak coolant requirement of 7kW which rapidly falls to 1kW. It is likely that a readily available commercial unit of 10kW would be used.

UNDERPINNING MODELLING

Preliminary modelling of the rate of waste freezing was performed in order to understand the projected rate of recovery for different design options and to establish sensitivity to waste and environmental process variables. Recognizing the tank wall temperature constraint (see operational constraints section), a range of design/deployment refinements were included (including the use of pokers/pins to increase the proportion of lateral freezing) to maximize the rate of waste recovery whilst minimizing the temperature change in the direction of the tank wall. Further refinements to the deployment also included (i) the addition of thermal stand-offs (insulated or heated tips), which are not currently included in the model and would form part of further work and (ii) provision of heating of the tank wall as a measure to maintain the required tank wall temperature.

Providing such protection to the wall would allow faster freezing, and so reduce cycle times and accelerate residual solids recovery.

Model description

The model is based around a distributed energy balance over the sludge phase, with some additional assessment of the freeze plate itself. It is important that the changing properties of the sludge as it freezes are calculated and that the phase change is adequately represented. Heat is transferred through the sludge by a mixture of conduction and convection. For relatively well settled sludges such as those in the tanks, conduction dominates since the solid particles slow the free motion of fluids under these conditions. Small increases in heat transfer due to convection can be represented as increases in the effective thermal conductivity of the sludge.

The freeze grab model can assume either that there is a fixed coolant temperature such that the model calculates the cooling required to maintain the temperature or a fixed coolant supply with a variable coolant temperature at the cryohead. The fixed temperature has been used for design of the cryohead. The sludge volume is modelled as a three dimensional block using a series of partial differential equations to carry out a heat balance [3]. The sides of the sludge block are assumed to be at fixed, ambient temperature. This is considered to be a pessimistic assumption as the freeze grab is likely to reduce the temperature at the boundary. For initial design, two boundary conditions were considered for the tank wall: in the first the heat transfer across the wall was considered to be zero. This assumption will increase the freezing rate, but will also increase the rate of temperature drop at the wall. The second condition assumes there is a fixed temperature underneath the sludge block (either at the wall or within lower parts of the sludge if the freezing is well away from the wall). This case will slow the rate of cooling also keep the wall temperature higher.

Cryohead Operation

A wide range of cryohead designs and operational variables were studied. The variables considered included the depth of the sludge, thermal conductivity of the sludge, the solids concentration within the sludge, solute composition and temperature of the cryohead. The modelling predicted that for sludge with the anticipated thermal conductivity (3.9 W/mK), after 12 hours of freezing a thickness of ~ 0.1m of ice is formed on the cryo-face (Figure 6). The geometry of the cryo-face then dictates the mass of ice retained.

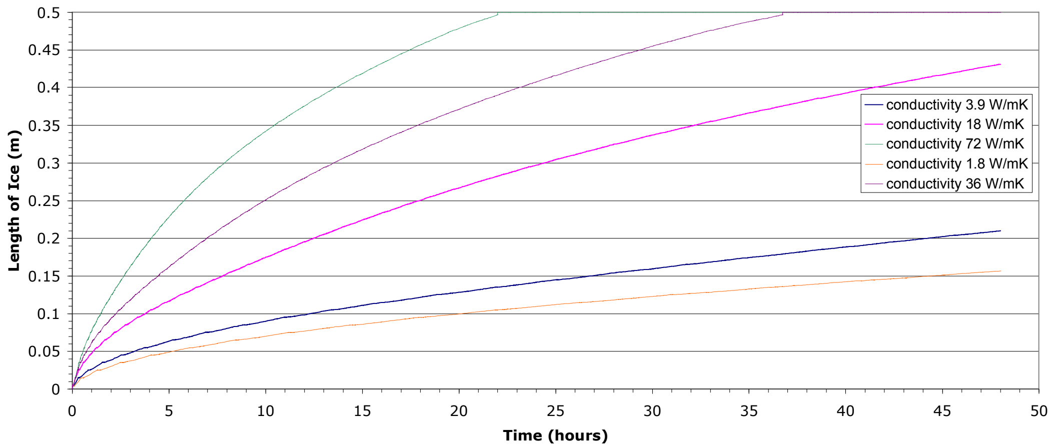

Fig.6. Base case model prediction for sludges with different thermal conductivity

Particular attention was paid to establishing how close the cryohead could be deployed to the tank wall without breaching the 21ºC tank wall temperature limit. Illustrative temperature profiles are provided below (i) for a flat plate design (Figure 7); and (ii) for a flat plate with additional protruding poker features (Figure 8). Protruding features are considered as they increase the rate of freezing, and also reduce the temperature (by virtue of the fact that more lateral freezing occurs). The freezing occurs in the X direction with 0 m representing the cryo-face and 0.2 m representing the tank wall.

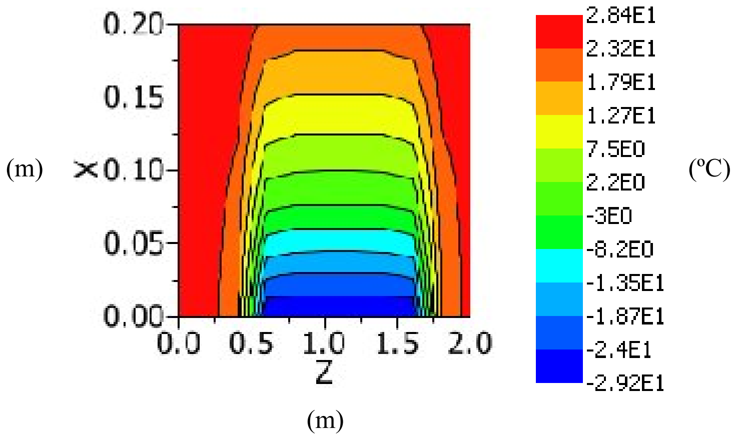

Fig.7. Temperature profile when tank wall temperature constraint is reached with isothermal tank wall heated to 30ºC with coolant at -30ºC

In Figure 7 it can be seen that for an unrefined geometry (i.e. without the benefit of pokers/prongs and thermal stand-offs) that it is possible to freeze within ~12 cm of the tank wall without breaching the temperature constraint. From the studies conducted so far it seems likely that repeating the work to assess smaller depths again e.g. reducing from 20cm sludge thickness to 12cm will allow removal of the sludge very close to the wall although as the depth decreases the amount removed per deployment will reduce.

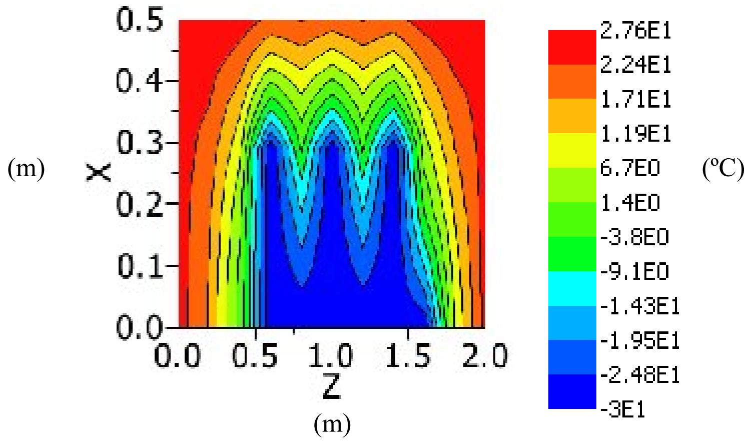

Fig.8. Temperature profile of 30ºC isothermal system with coolant at -30ºC when wall temperature constraint is reached

Figure 8 illustrates that when poker/prong features are included, ice is formed between the prongs. The case illustrated in the modeling presented here is not optimized with respect to geometry and additional features (such as thermally insulated tips – Figure 9).

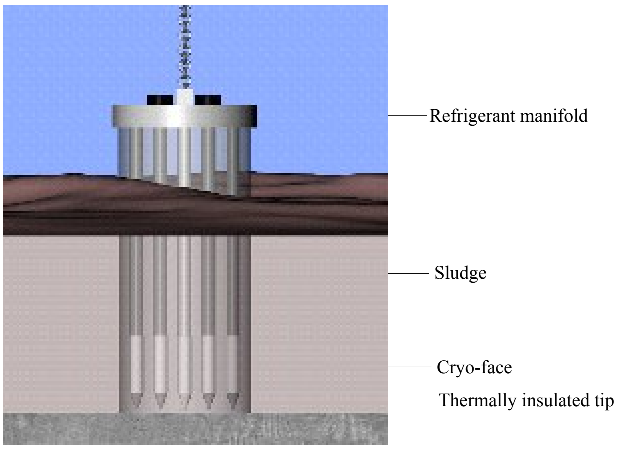

Fig. 9. Illustration of refined design

The modelling results supported the conclusion that the success criteria can be met when the associated operating constraints are taken into consideration.

DISCUSSION

Cryograb has been used in a variety of related nuclear and non-nuclear sludge recovery operations [3]. A principal advantage of the technology over other grab or hydraulic methods is the ‘complete’ nature of the grab achieved, coupled with minimal disturbance of the waste which prevents re-distribution of the waste during retrieval (so-called “chasing the waste”).

Prior successful deployments for the recovery of sludge and items in nuclear environments would suggest a high Technology Readiness Level (TRL) for both the cryograb technology and deployment system. Deployment in certain tanks at the Savannah River complex requires particular attention to maintenance of the temperature of the tank wall to prevent damage to tank infrastructure and hence a conservative TRL is currently claimed. Modelling has indicated that it is possible to satisfy this criteria, however a more conservative approach to waste recovery (relative to previous deployments) is required and additional engineering features may be required (e.g. thermally insulated stand-offs – Figure 9).

Based on a review of the deployment challenge, which defines the maximum dimensions of the cryo-head to 8 inch diameter or less, conservative assumptions on the mass of material that can be lifted per retrieval, and associated freeze rate modeling it has been estimated that recovery of up to 8 x 2 foot mounds and preparations to redeploy could be achieved within the 3 month window. The projected rate meets the defined in the success criteria and hence it is possible to conclude that deployment of cryograb present a viable option to support the recovery of residual sludge. Further work with inactive waste simulant is now required to underpin the projected recovery rate and temperature gradients through the waste.

CONCLUSIONS

Cryograb has been effectively employed for the recovery of nuclear and non-nuclear sludge. The principle advantage of the technology over other grab based technologies, and hydraulic methods is that waste re-distribution is minimized. This reduces the number of deployments required to recover waste.

Evidence to support a TRL of 2 has been demonstrated for the cryograb technology for the sample deployment under consideration. The sample deployment involved access through 8” gaps in risers with 35’ vertical reach and 12’ horizontal reach to recover 2’ residual mounds of sludge waste.

Adherence to a tank wall temperature constraint of 21ºC (applicable to certain tanks at Savannah River) requires the adoption of a more conservative grab approach and the adoption of additional engineering and control features. Modelling has demonstrated that freezing to within a few inches of the tank wall is possible. Further work with simulated waste is required to underpin model projections.

Cryograb provides a viable technology option to accelerate tank closure, specifically in cases where the presence of residual mounds of sludge are an issue and access is physically restricted.

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