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YUCCA MOUNTAIN NEAR-FIELD ENVIRONMENT CONSIDERATIONS FOR ENGINEERED BARRIER SYSTEM DESIGN AND PERFORMANCE

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INTRODUCTION

The United States Department of Energy (DOE) is investigating the suitability of Yucca Mountain (YM) as a potential site for the nation's first High-Level Nuclear Waste Repository. The site is located about 120 km northwest of Las Vegas, Nevada, in an area of uninhabited desert. Lawrence Livermore National Laboratory (LLNL) is a Project participant responsible for develop-ment of waste package (WP) and engineered barrier system (EBS) design concepts. This responsibility includes materials testing and selections, design criteria development, waste form characterizations, performance assessments, and Near-Field (NF) environment characterization. These areas of responsibility are inter-related and to a large extent depend on environmental conditions surrounding the EBS components.

The focus of this paper is to discuss what is currently known about the NF environment, particularly those aspects whose interaction with EBS components have significant impacts on the performance of the EBS and its ability to contribute to the isolation of radioactive waste. While this report will discuss NF environment as currently understood, this environment depends on many processes and design details that are not fully understood or determined at this time. Reference designs and design/processes assumptions are used to evaluate the expected NF environmental conditions. However, quantitative analysis of the actual NF environment must incorporate into the models information from EBS and repository design, operational practices and sequences, and performance assessment which are not available. Thus, this report will not focus on providing parameter values but rather on the processes that influence those parameters and on the impacts of those parameters on the performance of EBS components. Where values are given, they should be viewed as typical of YM as a whole but not necessarily representative of the actual NF environment conditions that surround the EBS components.

Major interactions between the NF environment and EBS components involve: the mechanical loading imposed on container materials and waste forms by the host rock environment; the amount of fluids and vapor or gases that contact the container or waste forms; and the chemical composition of those fluids and vapors. Interaction between the EBS components and NF environment is to a large extent dominated by the possible presence of liquid water. Corrosion mechanisms of most candidate container materials are almost exclusively aqueous, and leaching of most of the radionuclides is also almost exclusively dependent on the presence of liquid water as are transport mechanisms of the nuclides through the rock mass. The only nuclides that do not depend on water leaching and transport are the gaseous nuclides mainly associated with spent fuel (gaseous nuclides have been removed from the glass waste form and the only potential ones of concern are those generated slowly by radioactive decay). Thus, an assessment of the major environmental impacts on EBS performance can focus on the hydrologic aspects (particularly the liquid phase).

Favorable aspects of Yucca Mountain as a potential site relate to its arid nature and the sorptive properties of the rock materials. The arid environment results in unsaturated conditions at the proposed emplacement horizon. The major advantages of unsaturated conditions are that (without contact between liquid water and the WP) container corrosion, waste form leaching and transport mechanisms of radionuclides are minimized.

The major challenge of an unsaturated site is not due to anticipated poor performance as a potential repository site. Indeed, such a site is anticipated to perform better than a saturated site, providing all other aspects are comparable. The challenge is that the techniques available for characterizing unsaturated hydrology, particularly in fractured porous rock, and assessing waste isolation performance in that environment are neither well developed nor demonstrated. For saturated conditions the techniques are much better developed and there is a long history of application of the analytical tools, especially those related to porous media flow. For unsaturated conditions many of those tools or techniques are being developed. In addition, unsaturated flow is physically more complex. The balance between our ability to demonstrate or understand the hydrologic conditions and the site's performance is illustrated on Fig. 1. As can be seen, if a site is saturated our ability to demonstrate its performance is greater although its actual performance may not be as good as an unsaturated site.

The challenges in demonstrating performance or behavior are heightened by the time scale of the processes that must be assessed. Regulations require that radionuclides be essentially contained within the containers for 300 up to 1,000 years and that subsequent releases be at a very low rate for the next 9,000 years. The reliance on the waste container for the first 300 to 1,000 years requires corrosion rate and mechanism predictive capabilities and, thus, ability to predict water quantity and chemistry contacting the waste. The 9,000 year low release rate is currently assumed to be achievable due to very slow waste form leaching that is also a function of the quantity and chemistry of water contacting the waste.

Much of the work performed by LLNL has been to improve our ability to demonstrate the hydrologic performance. The predictive understanding of the hydrologic processes including perturbations over a 10,000 year time frame is unprecedented and must rely on mechanistic understanding.

Because the hydrologic aspects will need significant technical discussion and acceptance, this paper will deal with a portion of those more challenging issues related to hydrology and some aspects of mutual influence between geochemistry and hydrology. Discussion of other aspects including geochemical aspects can be found in various references.[1,2,3]

DISCUSSION

Yucca Mountain consists of a series of volcanic tuffs. The proposed repository horizon is in a densely welded-fractured tuff unit called the Topopah Springs tuff (Tpt). Estimates of fracture density (based on core analyses) range from 20-42 fractures/m^3.[4,5] There are questions regarding the estimating techniques used, but it is clear that the welded tuff is highly fractured.[6] Evaluations of in situ permeability data from boreholes (based on the cubic law) indicate that even with as many as 100 fractures/m^3, the fracture apertures required for the observed permeabilities would range from 43 to 127 μm.[7] As will be discussed in greater detail, this fracturing has a major impact on the hydrologic response of the NF environment.

Based on samples taken from surface coring, the Tpt is expected to have 65% saturation (±19%) and a porosity of 15%.[8,9] Air in the unfilled voids is expected to be moist (100% humidity). Overall infiltration is expected to be very low. Flux estimates within the repository horizon range from .5 to 1.0 mm/yr, possibly upward due to vapor transport from the underlying saturated zone.[10] "The amount of net infiltration that remains in the unsaturated zone is small when averaged over long time periods, because the estimated rate of potential evapotranspiration (1600 mm/yr) greatly exceeds the estimated average precipitation (150 mm/yr)..."[5]

Because of very fine matrix and unsaturated conditions, there is a high suction (matric) potential which will hold water within the pores. At lower saturations water is held in smaller pores or portions of the pores as isolated or discontinuous pore water. Only as the degree of saturation increases sufficiently to connect water between pores to form a continuous flow path can water flow through the matrix. Current estimates are that the majority of the pore water will not travel through the pores (porous media flow) until somewhere around 80% saturation is reached because the relative hydraulic conductivity remains low until nearly 80-90% saturation.[11] Also, water will not flow from the pores in the matrix to enter openings with diameters larger than the pores themselves since the capillary forces are an inverse function of the pore or opening diameter. This is the basis of one of the design features of the WP emplacement configurations, a capillary barrier between the WP and the borehole wall so that no pore water should contact WPs.

As long as conditions remain unsaturated and the capillary barrier remains (no WP contact with borehole walls or sloughing of rock into boreholes), there is no mechanism to allow water from the pores to contact the WPs. Thus, fracture flow is the only credible mechanism to bring water into contact with the WPs. Fracture flow, under capillary equilibrium between matrix and fractures, is not expected unless the matrix adjoining the fracture is nearly saturated (about 95%). A detailed theory addressing matrix versus fracture flow has been developed which indicates that fracture flow occurs either under conditions where the matrix becomes nearly saturated or for relatively large apertures. Application of that theory indicates that, for the strata being evaluated as a possible repository horizon, there is minimal possibility of fracture dominated flow.[12] It is possible for fracture flow to occur in response to infiltration pulses that exceed the rate of imbibition, and this fracture flow can occur to considerable depths.[13,14,15] It has also been shown that once the source of water is taken away, any fracture flow that had resulted from the rapid infiltration pulse would be imbibed such that the next pulse would not see the previous pulse. Therefore, fracture flow is not additive in that successive pulses cannot push the previous pulses further down the fracture. It is only when the duration as well as quantity of infiltration is much greater than expected that water can flow along fractures for the significant distances necessary to contact the WPs. Analyses indicate that standing water that remains for 48 hours at the entrance of a fracture may migrate as much as 100 m in a 100 micron fracture before the pulse is dissipated.[11,14]

As mentioned, fracture flow ceases as soon as the source is removed and the water that is in the fracture is imbibed into the matrix. This implies that fracture flow, if any did occur, extending to the WP horizon would be relatively short-lived. Preliminary analyses indicate that this may be consistent with observations of multiple levels of ^{36}Cl from core samples taken at YM.[16] The multiple levels may indicate that the pulses carrying ^{36}Cl may have extended to some depths but ceased as soon as the precipitation event ceased. The amount of water that could contact the WPs, even under the extreme conditions sufficient to allow fracture flow to extend to the repository horizon depths, would be limited both in quantity and contact time. Furthermore, if water were able to contact waste forms and leach radionuclides it would be imbibed into the matrix as soon as the source of the pulse was removed. A detailed theory addressing matrix versus fracture flow has been developed which indicates that fracture flow occurs under conditions where the matrix becomes nearly saturated or where there are relatively large apertures.[11]

The preceding hydrologic discussion assumes unsaturated conditions similar to current site conditions. It does not address changes that can take place during the 10,000 year time frame nor changes in response to the emplacement of waste. Waste packages will interact with a NF environment that will be altered from the original conditions by several factors including: construction activities, introduction of materials associated with repository and waste packages, heat generated by emplacement of waste, and the radiation associated with the waste. All of these can cause chemical changes, possible mineralogical or basic rock mass property changes, as well as hydrologic changes.

The evaluation of the potential for climate or seismic (if possible) changes to occur that are sufficient to change the unsaturated character of the site is not complete. Therefore, the impact on the WP is left for future studies. However, it is possible to evaluate the impact of construction and waste emplacement on the environmental conditions of the NF. It is also possible to make projections of the results of cooldown and return of moisture. To do this a conceptualization of the NF environment with time; i.e., moisture conditions, is presented in Fig. 2. This figure starts with the expected ambient conditions discussed above of 65% ± 19% saturation. The conceptualization of the ambient conditions is based on expectation that saturation conditions are normally distributed.

During construction, ventilation of drifts will remove moisture. Borehole drilling will also likely change moisture conditions. If water is used in drilling, the saturation of the surrounding rock will increase, and fracture flow away from the boreholes will occur. If the drilling fluid is air or air mist, the rock saturation will increase only if the partial pressure of the air/mist is greater than the partial pressure of the moisture held in the pores by the suction potential. If the air is dry, the saturation will be lowered around the borehole. Current plans call for dry drilling and construction so that the likely emplacement environment will be dryer than the ambient or current state. This is shown conceptually in Fig. 2 as the construction case. Drying by ventilation and drilling will not be spatially uniform; therefore, the distribution will be broader, and the mean saturation will be less that the current 65%. Information is insufficient to allow analyses of actual distribution at this time, and, therefore, the conceptual distribution is subject to change.

After waste emplacement, thermal loading will drive moisture away from boreholes. The thermal loading will be changing with time since radioactivity, the source of the heat, decays with time. Calculations performed for spent fuel 8.5 years out of reactor core (Fig. 3) indicate that after 100 years, heat generation is only about 22% of the original emplacement value. However, the effects of heating last much longer than 100 years. Fig. 4 shows the location of the boiling point isotherm with time. As is apparent, rock will be above the boiling point to considerable distances from the WP. It should be noted that these calculations are made based on young, and therefore, hot waste.

The conceptualization of post-emplacement saturation distribution is very much skewed towards zero values (Fig. 2) to reflect the very dry conditions that result from heating of the rock mass. The distribution shows a spike somewhat above zero to account for the possibility of some small percentage of the WPs, particularly those in the outer regions of the repository, to have some moisture in the NF. However, based on the temperature calculations referred to above, it is unlikely that any of the

WPs will have moisture in their associated NF environments and the conceptualization is likely overly conservative. Given the temperature distribution shown it is very likely that the NF environment will continue to be essentially dry for at least 300 years.

Since matric potential is a function of the degree of saturation, during the period when the NF is very dry there is very little possibility for pore water to contact the WPs. In addition, fracture flow is influenced by the imbibition of water into the matrix. As long as the degree of saturation is very low, the rock has a very large capacity to imbibe water into the matrix so that the potential for fracture flow to impact the WPs is unlikely. The dried out zone will act as a sponge to hold in the matrix any water that enters the dried out zone. The volume of rock dried out is very large (depending on the areal power density and thermal characteristics of the disposed waste), and thus imbibition capacity is very large.

After the waste and rock begins to cool sufficiently for water to return, moisture conditions for the coldest packages will tend towards ambient, and, therefore, the saturation for the ensemble of all containers will have a more broadly spread distribution with a mean only slightly increased above the 300 year value. It is impossible to determine how rapidly this cooling will take place, since the temperature history is very much a function of design and operations of the repository (which are yet to be determined). Therefore, the rate of change in the saturation distribution cannot be determined at this time. It has been demonstrated during field studies that the rewetting takes much longer than the drying cycle.[17] It is not determined at this time whether this was caused by the thermally overdriven nature of the field test or if it is independent of the thermal overdriving. If it is independent, then the time for saturation levels to return to ambient would be very long. Regardless, given the long period of elevated borehole wall temperatures (as much as 1000 years above or near to the boiling point) it is likely that the NF environment will remain dryer than ambient well beyond 1000 years. This is shown conceptually on Fig. 2.

Following total dissipation of the thermal pulse and after sufficient time has elapsed to allow moisture redistribution to occur over the entire altered zone, a hydrologic equilibrium condition will be reached. This distribution will match original ambient distribution only if there is sufficient moisture in the system to replace that which had been removed by the construction and emplacement processes or that drained away from condensation zones. It is assumed that it will take considerably longer than 1,000 years for the moisture to be replaced. The distribution is shown conceptually in Fig. 2 assuming that 10,000 years after emplacement all temperature and hydrologic perturbations will have dissipated.

These concepts potentially have very positive implications for WP performance. If saturation conditions can be demonstrated to be similar to the conceptualization shown for the 1,000 year post-emplacement period, water is unlikely to come in contact with the container during the entire

1000 year containment period. Analyses indicate that even under the high flux conditions of 1.0 mm/yr, "At no time within the 2600 year time span . . . is . water able to enter the borehole . . ."[18] Since corrosion is a function of contact with water this would mean that the containers would be anticipated to have at least a 1,000 year lifetime. Furthermore, the sponge effect would inhibit the movement of any radionuclides that might escape the WPs. Since the conceptualization assumes that the altered zone is at least as dry as ambient until well after 1,000 years this would imply that radionuclides would not leave the altered zone for many thousands of years. Even at ambient conditions, matrix flow does not occur and fracture flow only occurs in pulses which are ultimately imbibed into the matrix along with any radionuclides that are carried in solution. If there are any radionuclides carried as colloids they may filter out on the walls of fractures, but would not be moved further unless a more significant pulse occurred to pluck the colloidal sized materials off of the fracture surfaces. Even in this case, once the colloids are dried these materials may form fracture coatings that are not readily picked up by subsequent water pulses.

WATER COMPOSITION

Water composition in the near-field environment is not currently well characterized; however, reasonable bounds can be placed on the chemistry. Water is expected to be dilute bicarbonate water. Water from a well (J-13) completed in the Tpt in a location where the unit is saturated has been used to estimate the water composition that may be expected. This water is not in total chemical equilibrium with Tpt but is not drastically different than water in equilibrium. Samples of pore water from the densely welded tuff are not available, although samples from the non-welded units have been obtained. It is anticipated that the water from the pores will be in local chemical equilibrium with the tuff.

Composition of the pore (or vadose) water has been the subject of lengthy discussions and proposed studies, especially concerning the challenges of sampling the vadose water for chemical analyses. From the perspective of the NF environment, it is very important to understand the vadose water chemistry. As the pore water is evaporated or boiled off by heat, there is potential for rock/water interaction and for salt deposition within the pores. The amount and composition of any salt deposit will be a function of the initial vadose water chemistry. After the heat dissipates and the rock begins to gain moisture again, that moisture will contact the salts left behind and could create a much more chemically aggressive fluid. However, the determination of actual vadose chemistry is probably not as critical for WP performance as many assume. Based on our understanding of the hydrology, there is very little likelihood that pore fluids will ever come in contact with the WPs unless the capillary barrier deteriorates with time or the containers fall to the side of the borehole. The water that may contact WPs would be water flowing through the fractures themselves. The major impact of pore fluids would be on chemistry of imbibed water with very little influence on water flowing in the fractures. Only the water in pores that are in direct contact with the fracture surfaces can influence the chemistry of water in fractures. Interior block pore water can influence the pore water chemistry along fracture surfaces by slow diffusive processes, but that would not impact individual pulses in the fractures. One aspect that is not currently understood is how much microfracturing may take place which would bring a larger percentage of water flowing through the fractures into contact with the pore fluids which may have been altered by the remaining salts. Unless there is a significant amount of microfracturing it is likely that much of the salt deposition will be in the interior of blocks where it will not influence the chemistry for water in fractures. It seems likely that the majority of the pore fluids will never actually contact the WPs and waste form, but may migrate within the matrix into positions where that chemistry could influence the radionuclide transport and retardation mechanisms.

As mentioned above, the most credible mechanism for contact between the WP and water is through fracture flow. Fracture flow, as long as the NF is dry, will only occur in response to significant pulses. Since the chemistry of the pulses is unknown, the extent to which pore water chemistry along the fractures will influence water chemistry of the fluids in the fracture is unknown. However, the water chemistry of fracture flow is likely to be dilute since only pulses resulting from intense long duration rainfall could reach the WP horizon. This intensity of precipitation would likely be nearly pure water and, since the water will not remain in the fracture once the source of the pulse is removed, there would not be long periods of time for geochemical interactions with the rock materials.

For this reason, it is judged that understanding the exact chemistry of the pore fluids is not necessary to assess the performance of the WPs. It is necessary to assess the radionuclide transport and retardation. It is important to characterize the general composition of the pore fluids since many processes and activities that take place during and after construction and waste emplacement will influence hydrologic conditions and can create localized perched conditions.[6] The chemistry of this water can be different than water that is percolating through the mountain.

Several hydrologic process during drying out of the near-field environment will interact with geochemical processes. In the regions where rock is dried the processes will be dominated by vapor flow and chemical exchanges associated with vapor flow (Fig. 5). The zone of rock where temperatures are elevated, but are below the boiling point, will be very active both hydrologically and geochemically with potential for very active dissolution and precipitation of minerals. In the portions of this region where temperatures are very near to boiling, the rock will be undergoing drying with dissolved minerals left in the pores of the rock. This zone will be fairly limited in extent, and the moisture removed as vapor will be redistributed just beyond this zone in a condensation halo where both saturation and temperatures are elevated above ambient

(Fig. 6). Laboratory work indicates that healing of fractures can occur in this region.[19] As water is removed, dissolved materials will be deposited in the throats of pores, particularly the smaller ones. This may result in fundamental changes of the matrix hydrologic properties; i.e., elevation of the matric potential beyond values due merely to reduction in saturation. If these salt deposits are irreversible, then permanent changes may take place in porosity as well as matric potential. Because the groundwater is expected to be very dilute, the magnitude of this phenomenon is likely to be small, and it is expected to be reversible.

In the condensation zone the water deposited as condensate will be essentially distilled and more chemically active than the pore fluids that are somewhat in equilibrium with the rock matrix. Because of the elevated temperatures, it is possible that this zone will be one of increased geochemical activity. Because this is not a stationary zone, that is the boiling and condensation fronts move out from the WPs with time until the temperatures begin to collapse, the time for geochemical reactions to take place may be limited. The extent or significance of the geochemical reactions are not currently fully understood.

CONCLUSIONS

Waste emplacement environment will be essentially dry for at least 300 years and will likely continue to be dry for more than 1000 years, both as a result of the unsaturated ambient conditions and as a result of thermally induced drying. As the heat generated by the waste dissipates, there will be a gradual return to ambient (65%) saturation. Dry conditions are favorable to the waste container and waste form performance since corrosion by liquid water is the most aggressive environment for the container and since leaching by liquid water is the most likely mechanism for release and transport of radionuclides from the EBS.

Chemical changes may take place as a result of the drying process. These chemical changes may result in permanent changes in hydrologic properties. Specifically, porosity, hydraulic conductivity, and matric potential may be altered due to deposition of minerals within the pores as a result of drying. Also, fracture healing may result from mineral dissolution/deposition associated with heating/drying processes which would impact the potential for fracture flow.

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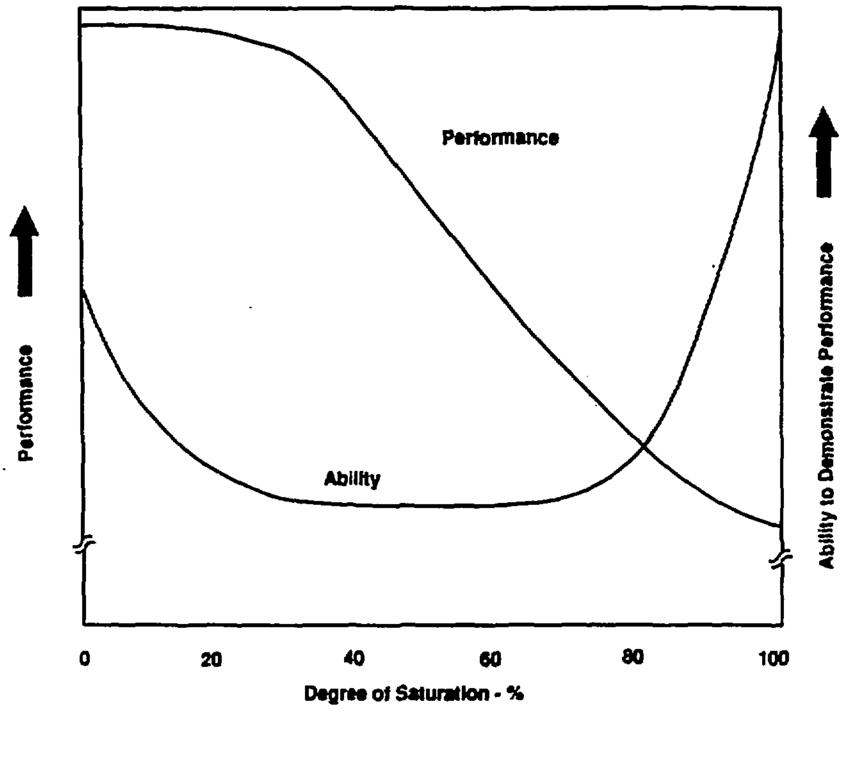

Figure 1. Site Hydrologic Characteristics vs Ability to Characterize or Demonstrate

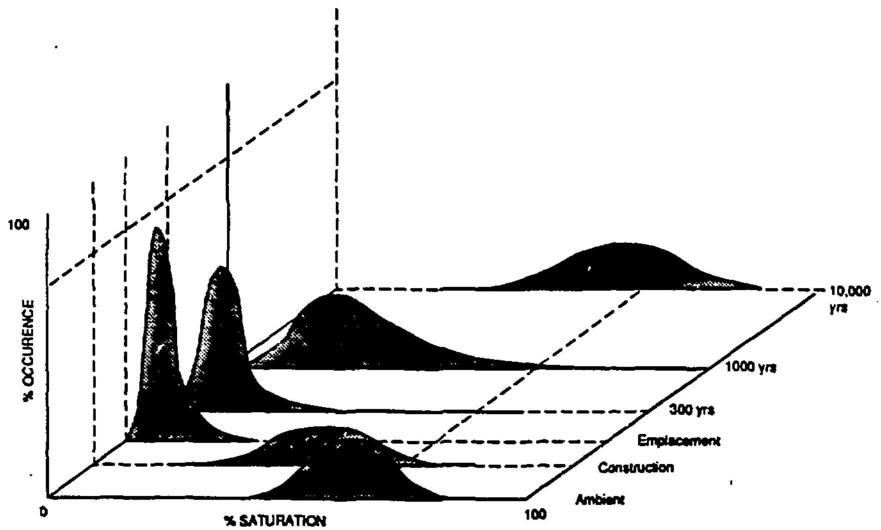

Figure 2. Saturation Distribution with Time

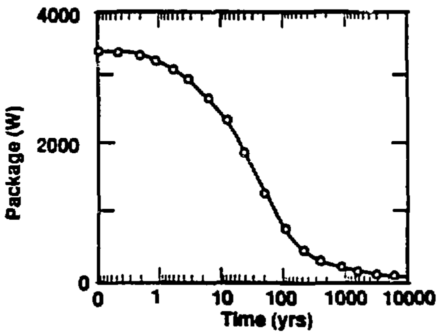

Figure 3. Power vs Time for Typical (8.8 yr old) Spent Fuel Package

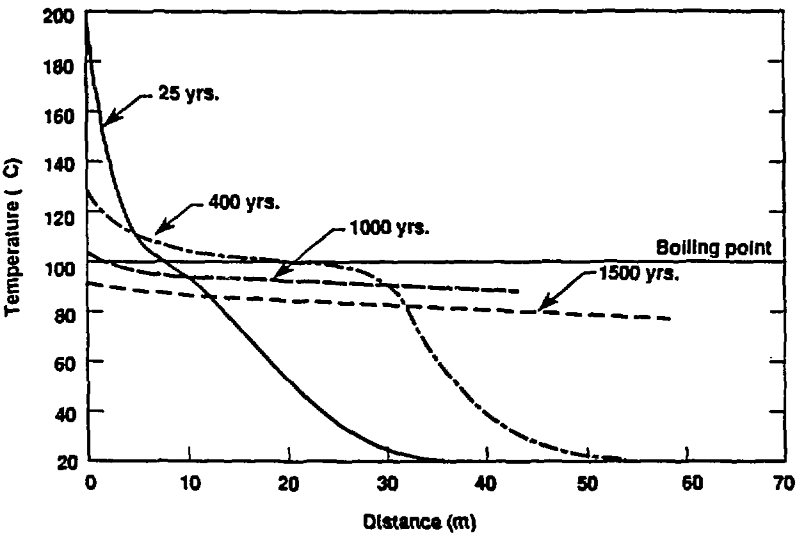

Figure 4. Temperature Profiles with distance

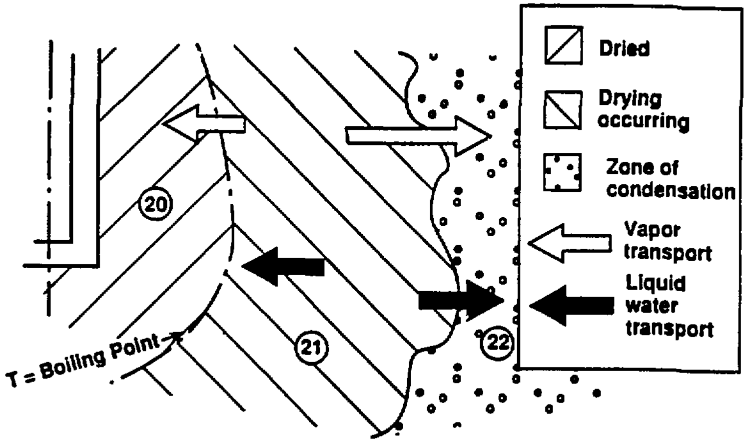

Figure 5. Environmental conditions zonation around waste emplacement

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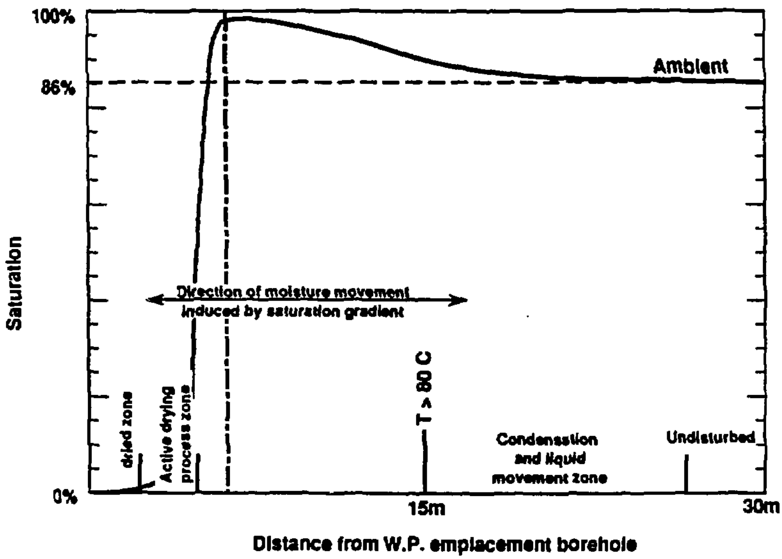

Figure 6. Saturation Conditions surrounding Waste Emplacement 25 years after emplacement