LA-UR-92- 654

LA-UR--92-654

DE92 008491

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SUBMITTED TO: The Waste Management '92 Conference
March 1-5, 1992
Tucson, AZ

Los Alamos

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SAFETY SUPPORT FOR HYDROGEN REANALYSIS OF WASTE TANK 101-SY

by

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ABSTRACT

Tank 101-SY, a double-shell tank on the Hanford SY high-level waste tank farm, has periodic releases of large volumes of gas. The released gas contains hydrogen (a fuel), nitrous oxide (a strong oxidizer), and other gases. These gases are intimately mixed, and therefore, it is very difficult to reduce the potential for a hydrogen combustion event. The safety issue is hydrogen gas exceeding one-quarter of the Lower Flammability Limit during these periodic releases. The Department of Energy Office of Environmental Restoration and Waste Management requested Los Alamos and Brookhaven National Laboratories to perform a reanalysis of a postulated hydrogen combustion event in Tank 101-SY. This paper provides the results of this work.

The results of this analysis are similar to the Westinghouse Hanford Company results with slightly higher pressures and larger releases. The results given here are believed to be conservative in that the pressures are higher and the radiological releases are larger than what would be produced by a best-estimate analysis.

INTRODUCTION

This paper discusses hydrogen reanalysis work performed for the Hanford high-level waste (HLW) tanks for the Department of Energy (DOE) Office of Environmental Restoration and Waste Management (EM). The Los Alamos National Laboratory and the Brookhaven National Laboratory (BNL) developed a joint program to perform the reanalysis; the analysis and its results are discussed here. An Unreviewed Safety Question was determined because of hydrogen gas generation and holdup in the waste of Waste Tank 241-SY-101 (referred to as Tank 101-SY). Tank 101-SY is one of 23 HLW tanks [5 double-shell tanks (DSTs) and 18 single-shell tanks (SSTs)] identified as having hydrogen gas generation.

Tank 101-SY, a DST, produces the largest amount of release gas. The release gas is generated in the slurry layer near the tank bottom and is released in an uncontrolled episodic venting. Release gas from Tank 101-SY contains hydrogen (the fuel), nitrous oxide (a strong oxidizing agent), and other gases that are mixed intimately. This results in a gas mixture that can be combustible even in the absence of air, thus making a hydrogen combustion event more difficult to mitigate. The interconnection of Tanks 101-SY, 102-SY, and 103-SY and the ventilation system (Fig. 1) affects the results of the safety analysis and has been included in the reanalysis methodology.

Currently, the processes of gas build-up and periodic release are not fully resolved. The best models at the time the reanalysis were performed assumed that the gas is generated uniformly. However, the gas in the viscous slurry layer is retained within this layer until the void fraction increases to the point where a density inversion occurs. At this time, the slurry and overlying liquid layers invert, and the gas is released from the layer and rises to the underside of the crust layer. These releases occur approximately every 90 to 100 days. Early data from release events in Tank 101-SY indicated that it took some time (on the order of minutes) for the release gas to "break through" the crust, which was believed to be a salt cake with some strength. This model of gas release led to two different types of accident to be considered: under-the-crust hydrogen combustion and dome-space (the free space above the waste surface) hydrogen combustion. These two types of accidents required different analysis methodologies using different computer codes to be developed. The methodologies are discussed in the next section.

Recent data from the last three release events in Tank 101-SY provided new information that raises questions about the gas collection under the crust and the crust strength. This information will be incorporated in a set of best-estimate calculations currently being performed. The under-the-crust accident analysis is being reviewed and may be modified to be more consistent with currently available information.

REANALYSIS ANALYTICAL MODELING

A Rayleigh-Taylor instability model was developed to predict the largest gas release and was used in all the analyses The worst-case analyses were performed with 57% hydrogen, 43% nitrous oxide, and 0% other gases.

Two types of postulated accidents were analyzed: dome-space hydrogen combustion and under-the-crust hydrogen combustion. In developing these analysis models, several factors were considered: the phenomena that need to be modeled, the complexity of data transfer between codes or models, and the accuracy that could be achieved in the results. This led to different analysis models for the dome-space and under-the-crust analyses. The analysis models have only two common elements: the maximum gas release model and the dispersion and dose model.

Dome-Space Reanalysis Analytical Models

Postulated dome-space hydrogen-combustion accidents assume the gas from a release event goes into the dome space (the free space above the liquid waste). An ignition source is assumed to exist at the time when the released gas has mixed with the air to form a worst-case hydrogen-combustion event. This is a more probable accident and represents a less severe environment for the tank structure than the postulated under-the-crust accident. Tank 101-SY has a ventilation system that affects the amount of mixing and limits the hydrogen concentration in the dome space. The ventilation system and its connection to Tanks 102-SY and 103-SY is considered in the analysis.

Three major computer codes were used to perform these analyses: the Hydrogen Mixing Study (HMS) code linked to the Transient Reactor Analysis Code (TRAC) and AI-RISK for the dispersion and dose analysis. The HMS/TRAC analysis modeled the injection and mixing of the release gas within the dome-space atmosphere. The combustion calculation was performed when a worst-case mixture is reached in the model. A crust-burn model was used to determine if crust ignition occurs and the extent that crust combustion would occur. The radioactivity source term in the dome-space atmosphere considers both the crust burning and convective pickup from the crust. In analyzing leakage to the atmosphere, three paths were considered: leakage through tank penetrations that are inlets under normal conditions, flow through the ventilation system [with high-efficiency particulate air (HEPA) filter failures], and releases from potential failures in risers and the tank itself. The structural response was analyzed by Brookhaven (1). The release of radionuclides to the atmosphere was determined with TRAC. The AI-RISK model used the calculated atmospheric release and a dispersion model coupled with the Hanford site meteorology to predict on-site doses for the closest facilities and off-site doses at the site boundaries.

Under-the-Crust Reanalysis Analytical Model

Postulated under-the-crust hydrogen-combustion accidents are characterized by a large volume of gas that is released from the slurry layer and collects under the crust for a short time (on the order of a few minutes) before flowing into the dome space. The under-the-crust analysis addressed the time before the gas flows to the dome space. As noted earlier, recent data from the last three gas release events provide more detail on the under-the-crust gas accumulation and crust properties (1). These new data have shown that the time the gas is under the crust may be very short and that the crust is not a hard salt cake as first believed. Under-the-crust combustions are currently under reinvestigation to provide a more realistic accident analysis

The analysis model for an under-the-crust accident is conceptually similar to the analytical model used to analyze combustion in the dome space. The major differences in the phenomena that must be modeled involve the dynamics of the crust as a result of hydrogen combustion below it and the generation of a radioactive source term resulting from the crust breakup. These differences are a result of the fast pressure loading of the crust. Two mechanisms for crust fracture exist: impulse loading from the gas combustion below the crust and impact with the upper part of the tank dome. These breakup mechanisms are treated independently in the analysis and result in a particle-size distribution for the radioactive source term in the tank. These fracture mechanisms generate a much larger radioactive source term in the dome space for this accident than is calculated for a dome-space hydrogen-combustion accident. Transport of the aerosol from the tank is modeled using TRAC. In this case, no aerosol contribution from crust combustion was included. As in the dome-space hydrogen-combustion analysis, AI-RISK was used to calculate both the on-site and off-site doses.

The hydrodynamics computer code MESA (2) was used to model the crust motion and deformation, to determine the resulting structural loading, and to estimate gross structural damage. Correlations of experimental data are used to predict the breakup of the crust into particles from both the pressure loading from below and from dome impacts. AI-RISK then was used to calculate the doses.

STRUCTURAL ANALYSIS APPROACH

The primary objective of the structural analysis was to determine whether the tank structures can safely withstand the postulated accident loads. If so, there will be small releases of radionuclides as a result of the accident. If not, the analysis should define the nature of the structural failure modes so that the release of radioactive materials and overall consequences of the accident can be estimated based on the leakage area.

Because the loads resulting from the accident scenarios are high, the tank structure will undergo a significant amount of plastic deformation for certain loads. This will change the geometry of the tanks and increase the interior volume, which in turn can reduce the estimated loads (e.g., hydrogen burn in the dome space). Therefore, the structural analysis also should identify any source that will alter the estimated pressure loads so that Los Alamos can recalculate the loads and BNL can perform the structural analysis with the revised loads.

Because of the complexities involved in the failure analysis of reinforced concrete/steel liner of the DSTs as discussed above, the approach adopted by BNL was to start with a simple conservative model and gradually add complexities on a step-by-step basis until a realistic (but still manageable) model was achieved. This approach allows the analyst to very and adjust the model at every step and compare the results as the model becomes more and more complex. This provides confidence in the results, especially for the subsequent, more complex models with additional load types

The initial model considered for the analysis was the primary steel tank without the secondary concrete structure. This model was analyzed considering only the membrane action of the steel shell elements. In addition, the tank was analyzed by constructing two-dimensional (2D) and three-dimensional (3D) finite-element models.

The second model considered for the analysis consisted of the primary steel tank, secondary steel liner, and reinforcing steel in concrete. The concrete itself was not included in the model. The analysis was performed with 2D and 3D finite-element models, and both small and large deflection options were used.

The third model represented the complete structure and included the steel tank, secondary steel liner, and concrete tank, including the reinforcing steel. A 2D model was constructed to analyze the structure.

All of the analyses were performed for static loads considering both nominal (i.e., lower bound) and mean material stress-strain values. The dynamic effect of pressure loads was addressed separately. Only the important and final analysis models and results are discussed in this paper.

101-SY REANALYSIS RESULTS

The initial conditions for the analyses were defined by a release gas composition and volume, which together give the mass of reactants. The composition of the release gas used in all of the analyses was 57% H_2 and 43% N_2O, with no diluents. This composition was dictated by concerns over the uncertainties in the method used to convert mass spectrometer analyses of exhaust gas to release gas composition. This problem has been reduced considerably since the reanalysis was started, and the composition used here now is considered extremely conservative. The volume of gas was calculated using a Rayleigh-Taylor instability model. It was assumed that gas generated in the lower slurry layer is retained until a density-driven instability occurs and the layers invert.

Based on measured temperature and density profiles, the model predicted a maximum release of 255 m^3 (9000 ft^3) at the conditions in the dome. This yields a total mass of 200 kg (440 lb), 11.2 kg (24.6 lb) of hydrogen and 189 kg (416 lb) of nitrous oxide. These values were used for both the postulated dome-space and under-the-crust hydrogen-combustion accidents.

Dome-Space Hydrogen-Combustion Analysis Results

The primary computational tool for the analysis was the code HMS/TRAC. HMS/TRAC is a coupled version of the individual codes HMS (Hydrogen Mixing Studies) and TRAC (Transient Reactor Analysis Code) (3). HMS was used to model the release of gas into the vapor space, the subsequent mixing, and combustion. The network flow capability of TRAC was used to model the ventilation system associated with Tank Farm SY.

The effects of the ventilation system and flame speed on peak pressure in the dome were studied. The effect of ventilation system failure was examined by first assuming that the tank was sealed. In a second calculation, the ventilation system was assumed to fail at the time of ignition. In the latter case, the combustible inventory was reduced somewhat. In both cases, combustion began simultaneously in all computational cells, which maximizes the pressure rise. More realistic calculations were performed with the ventilation system functioning normally and with a flame propagation model. Ignition occurred at the outer edge of the crust and at the end of the injection phase, when the gas inventory is a maximum. The peak pressure and temperature for all three cases are given in Table 1.

The structural analysis of Tank 101-SY was performed by constructing three models and adding complexities at each step as discussed earlier. The respective finite-element models were generated using the information available in Hanford drawings, specifications, and reports. All three models were analyzed for static loads, and the results such as strain and deformation because of varying internal pressure were provided for each model.

The basic behavior of the tank was consistent in all three models. The main weak part in the structure was the bottom knuckle region of the tank, and the primary resistance against uplifting was derived from the friction of surrounding soil. In Model 1, without the weight of concrete and overburden soil, the dome top moved upward freely, mainly as a result of bending of the bottom knuckle. In Models 2 and 3, the uplifting was delayed until the dead load was overcome by the upward pressure. The uplifting was delayed further when the resistance resulting from soil friction was included in the analysis. All models showed logically consistent behavior, demonstrating confidence in the modeling and computer codes. By comparing the results for all three models and considering the more realistic modeling, including the dead load, weight of overburden soil and soil friction, and mean steel strength, it can be concluded that the maximum vertical displacement of the dome top and strain at the primary tank bottom knuckle region will be limited to 16 in. and 2.7%, respectively, at an internal static pressure of 57 psig. These results compare favorably with the results published by Westinghouse Hanford Company. They reported a maximum vertical dome displacement of 35 in. without soil friction and 14 in. with soil friction at 60 psig.

Bending in the primary tank knuckle region produces both tensile and compressive stresses. Moreover, there is no source of strain concentration in this region. Therefore, no liner tearing is expected at this strain level. The knuckle plate is welded to a thinner wall plate. However, this transition is sufficiently away from the corner so that the meridional strain is reduced significantly. The penetrations through the dome can cause a large strain concentration. However, the strain level in the dome is low (< 6%) and not expected to be a threat. However, the vertical displacement of the tank is rather high and could be a concern for the integrity of the transfer piping and risers that cannot move freely. This requires further investigation; the conclusions are preliminary at this time. Currently, a damage study at locations with high strains and an investigation of possible damage to transfer piping and penetrations are under way.

The above-crust burn results in a single pressure pulse. The dynamic effect of the accident pressure was addressed by performing a modal analysis. Capacities and dynamic loads were compared to characterize the structural integrity of the tank because of the dynamic loads. This resulted inan attempt to define an equivalent static load resulting from this pressure pulse. The fundamental frequency is required for this purpose and was estimated at 7.1 Hz through a modal analysis. The amplified load at any particular time during the above-crust burn was computed by considering a 5% damped single-degree-of-freedom system with a fundamental frequency of 7.1 Hz. The amplification was negligible. To accommodate possible variation of the structural fundamental frequency (or the load frequency), a parametric study was performed with 5 and 9 Hz. Virtually, the amplified load at no time exceeded the peak load of approximately 55 psig. Therefore, the structural integrity of the tank as a result of the above-crust burn can be evaluated considering an equivalent internal static pressure of 55 psig.

Aerosols would be generated by crust burning and entrainment of dry crust particles by high-velocity gas during hydrogen combustion. The total amount suspended in the dome would be approximately 300 kg (660 lb). It was assumed that all of this material has a particle diameter of less than 10 µm. The amount of aerosol released from the tank was calculated using TRAC under the assumptions that there is no agglomeration or plateout in the tank or ventilation system, that there is no slip between the gas and solid phases, and that the HEPA filter has failed. With these very conservative assumptions, the total release was calculated to be 60 kg. Dispersion of the aerosol and the dose rate from the fallout were calculated with AI-RISK, a Gaussian plume model that tracks dispersion in five different sized bins. Both acute and long-term doses were calculated for on- and off-site locations. The results at the 95% level are given in Table II.

Under-the Crust Hydrogen Combustion Analysis Results

Postulated under-the-crust hydrogen-combustion accidents are characterized by the retention of a large volume of gas under the crust for a short time (on the order of a few minutes) before it flows to the dome space. An implicit assumption in this model is that the crust represents an appreciable barrier to upward gas flow and that it has sufficient strength to support itself. That is, the crust acts as a piston with the gas trapped below. Recent data have brought this picture of the crust in Tank 101-SY into serious question. We are evaluating the physical reasableness of this accident scenario.

The combustion process was represented as an instantaneous, constant-volume burn. This produces an initial pressure at the start of gas expansion of 161 MPa (236 psia) and a temperature of 4472 K (7592°F). Motion of the system as a result of the gas over-pressure also was calculated with the hydrodynamic code MESA (2), a multidimensional, multimaterial Eulerian computer code with material strength and failure models. The structure and tank contents were modeled in considerable detail. The expanding gase accelerates the crust upward into the top of the tank and produce significant deflections in the primary liner. The crust moves upward for 150 ms, compressing the gas trapped between it and accelerating the tank top. This is an efficient method for doing work on the tank. The upper structure continues to accelerate for 100 ms with a total deflection of approximately 1 m, indicating possible structural failure. The peak impulse to the dome was 94 $kN\text{-}s/m^2$ (13.64 $lb\text{-}s/in.^2$).

As discussed before, the internal pressure resulting from the below-crust burn is not uniform and is much higher than that from the above-crust burn. The impact loads are even higher (e.g., 21 bars on the dome surface and 43 bars on the bottom knuckle). The tank is expected to suffer potential catastrophic failure at such high pressures.

Aerosols are produced in this accident by the impulsive loads to the crust during its acceleration and by impact with the structure. Empirical correlations based on data for explosive dissemination of materials were developed to estimate the amount and size distribution of aerosols. The measure of explosive insult is the particle velocity in the material being aerosolized. Total aerosol production was 6023 kg with 42 kg less than 10 µm in diameter. The flow of this material from the tank was modeled using TRAC with initial conditions provided by MESA. For the same assumptions as in the dome-space burn (no slip, no plate-out, etc.), a release fraction of 0.37 was calculated. The corresponding dose rates calculated with AI-RISK are given in Table III.

These results are similar to those reported previously by Westinghouse Hanford Company. The differences can be explained in terms of the initial condition and modeling approaches. In general, our assumptions were more conservative, and the computer models we used were somewhat more detailed. It can be concluded that the worst-case progression of these accidents is well understood and that an upper limit on the consequences is available.

CONCLUSIONS

An independent reanalysis of the hazards associated with Tank 101-SY has been performed. Two postulated accidents, an above-crust burn in the dome vapor space with associated crust burning and a below-crust burn, were considered. In both cases, the total mass of the gaseous reactants was the same and diluents were assumed to be absent. The above-crust burn was analyzed using HMS-TRAC to calculate the mixing in the tank and the subsequent combustion. The crust burn was modeled using a discrete-ordinates model for radiation transport within the

the below-crust burn, the hydrodynamics were modeled using MESA, and the aerosol production was based on empirical correlations modified for this application. Aerosol transport from the tank was modeled in both cases with TRAC; dispersal and dose-rate calculations were performed with AI-RISK. The final dose off-site rates were 24 and 3 mrem for above- and below-crust burns, respectively. The dose for the above-crust burn is greater because of the associated crust combustion.

The structural analysis of Tank 101-SY was performed using three models with increasing complexities. In each case, the results where consistent with the modeling assumption and complexity. The dynamic effect of the accident pressure was addressed by performing a modal analysis capacities, and the dynamic loads were compared to characterize the structural integrity of the tank. The above-crust burn resulted in a single pressure pulse. We attempted to determine an equivalent static load resulting from this pressure pulse. The fundamental frequency is required for this purpose and was estimated at 7.1 Hz through a modal analysis. The under-crust burn scenario provided multiple pressure pulses and higher loads to both the dome and lower tank region. These were dealt with separately.

Several recommendations can be made based on this work. To reduce the conservatism in the analysis, better data are needed on crust properties and the kinetics of H_2-N_2O systems. These data will be available in the near future based on recent core sampling activities and planned experiments at the US Bureau of Mines. In addition, final agreement on the correct composition of the release gas should be reached and documented. The probability of an under-the-crust burn needs to be reexamined in light of the more recent in-tank observations. This also applies to a postulated combined burn. Finally, the effect of combining various conservative assumptions about material properties and physical models needs to be evaluated systematically.

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*A combined burn is a simultaneous above- and below-crust burn.

TABLE I. COMBUSTION RESULTS FOR DOME-SPACE BURN

Case	Pressure kPa	psia	Temperature K	°F
Sealed Tank	314	104.6	1879	2928
Volumetric Ignition	615	89.2	1920	2997
Flame Propagation	533	77.3	1720	2637

TABLE II. DOME-SPACE HYDROGEN BURN DOSES

Location	Distance (km)	EDE (rem)	Cancer Risk (lifetime)	Decontamination Factor
S-Plant	0.66 SE	2.9	3.2E-03	1
U-Plant	0.78 NE	2.4	2.6E-03	1
Hwy 240	3.9 SE	0.19	1.2E-04	1
Maximum Off-Site	13.8 WNW	0.024 2.3[a]	1.2E-03	1

[a]70-yr dose rate due to continued consumption of contaminated foodstuffs.

TABLE III. UNDER-THE -CRUST HYDROGEN BURN DOSES

Location	Distance (km)	EDE (rem)	Cancer Risk (lifetime)	Decontamination Factor
S-Plant	0.66 SE	1.7	3.3E-03	14.4
U-Plant	0.78 NE	1.1	3.1E-03	3.8
Hwy 240	3.9 SE	0.036	4.0E-04	1.0
Maximum Off-Site	13.8 WNW	0.003 1.4[a]	2.0E-03	1.0

[a]70-yr dose rate due to continued consumption of contaminated foodstuffs.

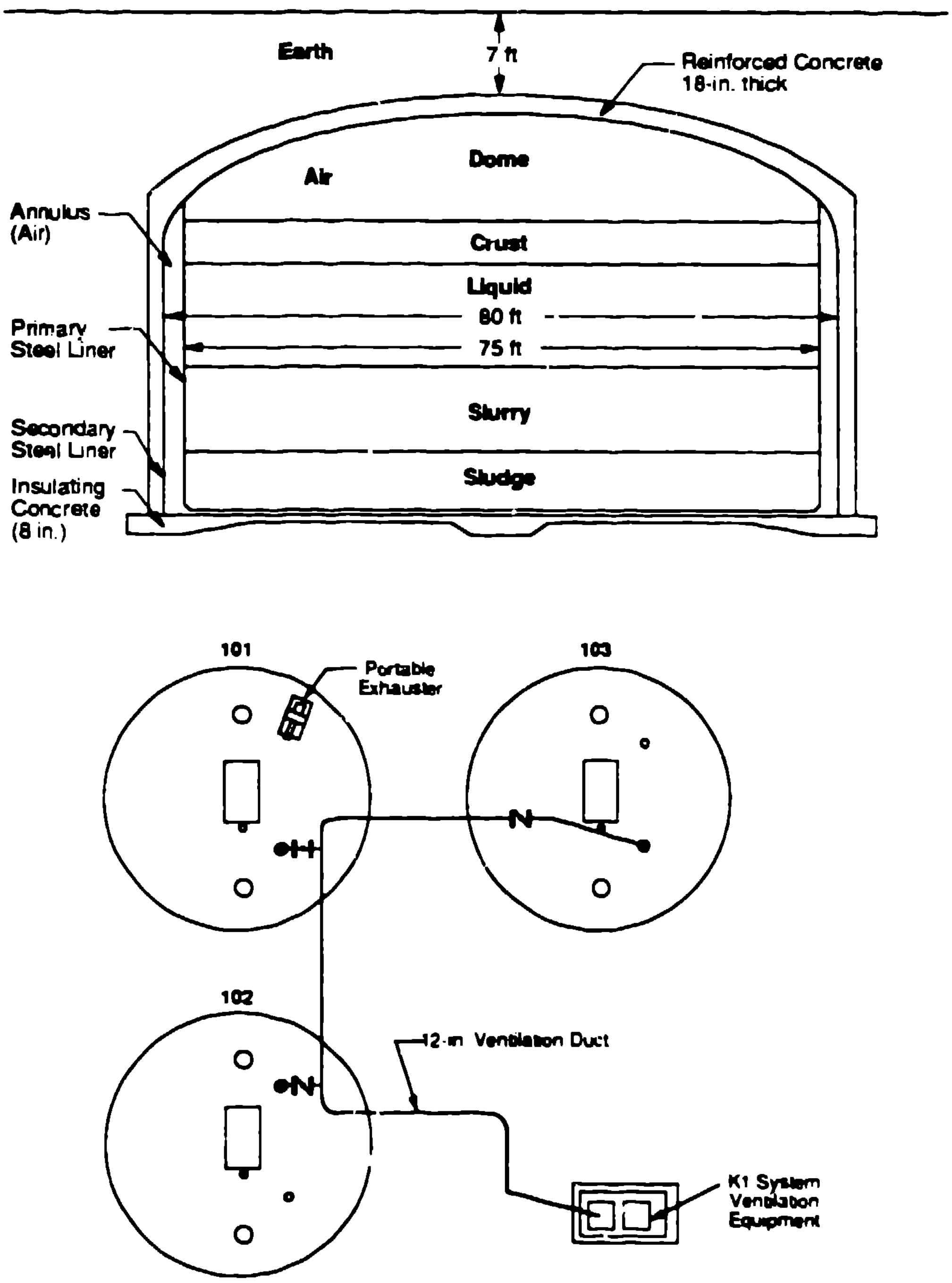

Fig. 1. Top: Tank 101 SY geometry and contents. Bottom: SY Tank farm.