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7. Abstract

Synthetic Hanford Waste form grout specimens were irradiated in a gamma field and gas generation and release were followed using pressure measurements and chemical analysis. Gas pressure activated liquid release was observed from only one of the irradiated specimens that was in a gamma field many times more intense than expected in a grout storage facility.

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**GAMMA IRRADIATION TEST REPORT OF SIMULATED GROUT SPECIMENS
FOR GAS GENERATION/LIQUID ADVECTION**

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Executive Summary

This report presents the results from an irradiation test performed on four specimens of grout that were fabricated from synthetic Double Shell Slurry Feed (DSSF) liquid waste. The objective was to investigate the radiolytic generation of gases and the potential for advective rejection of waste liquids from the grout matrix and to provide experimental information for the validation of the C-Cubed calculated model. It has been demonstrated that a number of gases can be formed within the grout due to radiolytic decomposition of various chemical components that make up the grout. This observation leads to the conjecture that the potential exists for the rejection of a portion of the 60 vol% free liquid from the grout matrix driven by pressurization by these gases.

It was found that, for the specimen geometries used in this test series, and for peak radiation dose accumulation rates on the order of 4 to 60 times of the initial rate expected in the grout vaults (300 Rads/hr), no liquid rejection was observed from 2% to 35% of the target exposure expected in the grout vaults ($1\text{E}+08$ Rads). When the irradiation rate exceeded the projected grout vault dose rate by a factor of 200 a small amount of liquid rejection was observed from one of two specimens that had received 20% more than the goal exposure. *Because of other experimental observations involving these two test specimens (gas release rates, gas and liquid compositions, etc.), it was speculated that the difference in behavior between these two seemingly identical specimens was due to unidentifiable specimen preparation dissimilarities.*

Because of the differences in the magnitudes of the relative radiation field strengths between this study and an actual grout vault, it is concluded that the potential for liquid rejection by internal gas pressurization from presently configured grout waste forms is very low for the expected conditions. However, because of the great difference in the volume, mass and surface area between the experimental and real cases, a more sound judgement about the probability of the occurrence of advective rejection of grout liquid awaits a thorough evaluation using the modeling codes.

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GAMMA IRRADIATION TEST OF SIMULATED GROUT SPECIMENS FOR GAS GENERATION/LIQUID ADVECTION

1.0 INTRODUCTION

1.1 Objective

The primary objective of these irradiation tests was to provide experimental information for the validation of a calculational model (originally called C-Cubed, later evolving into the REGAL code (Roblyer, 1993)) that describes the generation, release, retention and migration of radiolytically (and chemically) formed gases throughout the grout vault system. This model computes the generation within and the migration of gases from the grout matrix through the concrete walls of the vault, out the cracks at the top of the vault, down the outside walls to the catch basin, and then into the sump, where the potentials for the formation of explosive mixtures are calculated. This model also predicts a possibility for advective rejection of the liquid phase from the grout by internal pressurization by the radiolytically formed gases.

1.2 Background

Representative G-values describing the generation rates of various gases by radiolytic decomposition of grout components are required in the analytical model to assess the performance capabilities inherent to the design of the grout vault systems. Another experimental series, in addition to the subject of this report, is being carried out at Argonne National Laboratories (ANL) to obtain gas generation rate data from synthetic grout formulations. "Hot" gas generation rate tests are being conducted at Hanford using grout specimens fabricated from actual waste from tank 241-AP-102 that will provide the liquid for the Campaign 102 grout pour (Hendrickson et al. 1993). These radioactive-grout tests are designed to obtain supporting information to the ANL work. Another test series at Hanford is examining the potential for liquid rejection from the same synthetic grout used in this report by an externally applied gas and liquid pressure gradient (Crane, 1993).

The experiments that are the subject of this report complement the test series described above, and in addition, deal with an ancillary question related to the behavior of the grout as it accumulates radiolytically and chemically formed gases. This question is: "How does the liquid phase of the grout interact with these gases as the grout becomes pressurized?" A mechanism has been described (Roblyer 1993) that possesses the potential for initiating an advective rejection of the liquid from of the grout mass, driven by internal gas pressurization by the radiolytically produced gases in the liquid phase of the grout. Modeling studies of the full grout vault have shown that, for the expected gas generation rates, the advective release of liquids will probably not occur, but it is not clear whether the magnitude of the mechanism will remain at acceptable levels.

It was earlier estimated in the Test Plan for the present experimental series (Hinman, 1993) that for a G-value (number of all gas molecules produced for every 100 eV of deposited gamma energy) of 0.13 (the so-called "Reasonable Bounding Case"), a maximum internal pressurization of the grout of on the order of 50 atm (700 psi) could be achieved for a gamma exposure of $1\text{E}+08$ Rad. This is a conservative estimate, based upon the conservative assumption that no gas is released over the time period of the irradiation, and conservative

values for the solubility limits versus pressure for the gases that were assumed to be formed (H_2 , N_2O and O_2).

In addition, the results from the application of an early version of the C3 code (Roblyer, 1993) were reported in the Test Plan for the geometry and expected conditions of this irradiation test and some of the predictions are of interest as illustrative examples. The C3 code predicted that over 20% of the free liquid in the grout (about 50 to 60 vol% of the grout is free liquid) would be rejected by this gas pressurization for $G = 0.13$ molecules/100 eV and dose = $1E+08$ Rad. The C3 code even allowed some of the gases to diffuse out of the grout. This prediction does not necessarily apply to a grout vault because of the much higher rate of gamma exposure in these tests. It is not possible to determine the pressure at which the liquid is predicted to begin flowing from this modeling study. The significance of the gas pressure driven mechanism can only be resolved by experimental means.

This test series used relatively massive (compared to the ANL specimens), near-saturated synthetic grout specimens and subject them to a gamma radiation field in an attempt to determine the quantifying aspects for the potential liquid flow.

Experience with the waste tank, SY-101 (Pederson et al. 1993) has shown that gases can be formed in waste materials, through both chemical and radiolytic mechanisms. In addition, it is known that some chemical components that will be present in the grout feed materials (and waste tank liquids) can be decomposed into various gases by radiation. The main constituents in the grout waste-form that can form gaseous species are water, nitrates, nitrites, and to a lesser extent (because of the dilute nature of the species) the organic compounds of the liquid waste. Gases that can be derived from various components are H_2 and O_2 from the water, H_2 from the organic, and N_2O and N_2 from the nitrates and nitrites. All of these gases have been observed experimentally from simulated tank wastes in studies carried out at ANL (Meisel 1991) where synthetic waste solutions were subjected to gamma fields.

The chemistry of radiolytic decomposition of compounds is complex, but the qualitative aspects are clear: gases will be produced. The physical chemistry of the interaction between gases and liquids is also complex. Models for these interactions are being developed at Hanford (Roblyer 1993). As gases are generated in the grout, they are initially in solution within the liquid phase. All gases have a finite solubility in a liquid. The traditional way to phrase a description of the phenomenon is to say that the solubility (i.e., composition) of a gas in a liquid is a function of the pressure of the gas applied to the liquid, as expressed by Henry's Law. However, this conventional stratagem derives from gases entering the liquid from an external source. In the case of the grout, the gases are generated internally, and the phrasing should be inverted, i.e., the pressure exerted by a dissolved gas in equilibrium with the liquid is a function of composition of the gas in the liquid. The higher the concentration of gas in the liquid, the higher the external pressure required to keep it from nucleating into bubbles.

If gases stay in solution, an over-pressure that can be calculated from the aforementioned Henry's Law must be present. If the internal pressure of the grout becomes high enough, bubbles nucleate and grow by the addition of more gas molecules and by coalescence between bubbles. These physical processes must be accompanied by a volume increase. However, since the liquid is nearly incompressible, this results in a pressurization and a potential for flow of the liquid. About 50 to 60 volume per cent of the grout is free liquid, as measured from the weight losses during the drying of the grout.

In the grout, a fraction of the volume is taken up by air bubbles entrained during mixing and pouring (porosity). The internal porosity is available to accommodate the volume expansion and pressurization caused by gas bubble formation. However, internal

pressurization tends to cause flow of the mobile liquid components. The unchemically-bound liquid is contained in the labyrinth of the internal structure of the grout, and can potentially be moved if forced. This phenomenon is called liquid release by advection. As this process continues, it can become a two-phase flow of gas bubbles and liquid.

The gas molecules that remain dissolved in the liquid make up another mobile constituent within the grout mass. These molecules can move by diffusion in a chemical potential gradient, tending to alleviate the pressure buildup in the grout by a redistribution of the gas molecules from a high concentration (pressure) region to a lower concentration region. These gases can diffuse through the liquid to a free surface where they are released, relieving the internal pressure in the grout. The driving force for the diffusion of the gas atoms is the concentration (pressure) gradient in the liquid from the inner volume to a free surface where the release can occur. The free surface to which a gas molecule will diffuse is more likely to be an internal crack than one of the six large surfaces of the grout parallelepiped.

Modeling of these phenomena has led to the conclusion that the diffusion rate-limited release mechanism of gases competes with the advective release of gases where liquid is rejected. A generalized concept has been proposed (Roblyer 1993) of a liquid expulsion threshold for the gas generation rates, i.e., there exists a rate of radiolytic gas generation below which the gases can be released from the grout by diffusion through the liquid phase such that pressurization of the liquid is relieved to the point of never becoming high enough to push out the liquid. This threshold is dependent upon, among other things, the resistance to fluid flow by the grout matrix.

The experimental series that is the subject of this report was designed to examine the issue of advective liquid removal from the grout. Specimens of simulated grout were subjected to a range of gamma field intensities of well in excess of those encountered in the grout vault.

2.0 SYSTEM DESCRIPTION

Six cylindrical synthetic grout specimens were prepared at the 222-S Laboratory for the gamma irradiation experiment using simulated DSSF (Double Shell Slurry Feed) liquid waste and dry mix as described below. These specimens were poured into stainless steel containers (modified 2 liter beakers) that were 12.42 cm (4.89 inch) diameter and 14.92 cm (5.88 ± 0.10 in.) length. The specimen diameter allowed at least 5 mm clearance (centered) to the inside diameter of the outer capsule (i.e., 10 mm less than the capsule ID). The capsule requirements are discussed in Section 4.1.

The preparation of these specimens was coordinated with the production of smaller synthetic grout specimens for the ANL gas generation rate testing program (Powell 1994). Four of these six specimens were used in the gamma irradiation testing for liquid advection. Two of the specimens were not irradiated because of conflicts with other programs for space at the Gamma Facility and were used as blank tests to observe chemical gas production rates. The stainless steel containers into which the grout was poured minimized the evaporative loss of water from the surface of the specimen before and during irradiation, and tended to minimize the loss of generated gases from the grout by diffusion from the outer surfaces of the cylinders. The total volume of all six of these grout specimens was approximately 10.8 liters (660 in³), or 1.80 liters each.

The specimens were cured for 28 days under controlled conditions specified below. Each specimen was inserted into an outer sealed metal container fitted with a gas sampling valve, a liquid level measuring capability, a thermocouple, and a pressure transducer for real-time temperature and pressure monitoring. All hardware for the test apparatus was designed, fabricated and assembled in the 306E laboratory facilities (Feigenbutz, 1993).

2.1 SIMULATED LIQUID WASTE

The simulated liquid waste was mixed at the WHC 222-S Laboratory in accordance with the nominal composition in Table 1. A detailed description of the preparation of the liquid and its subsequent analysis at the fabrication of the grout specimens for this test series is contained in Powell, 1994. A quantity of liquid was mixed that contained enough material to fill the requirements for the six specimens specified in Section 3.0 later in this report. This composition derives from a combination of the Double Shell Slurry Feed (DSSF) waste liquids in two tanks that have been mixed together (Hendrickson et al. 1993). Tank 241-AN-106 contained about 88% (985,000 gallons) of the volume. This tank has been pumped into tank 241-AP-102 that contains a "heel" of liquid (135,000 gal) equivalent to about 12% of the final volume.

2.2 DRY BLEND

A dry blend was prepared at the 222-S Laboratory that contains enough material to fill the requirements for the number and size of specimens specified in Section 2.0 above. The composition of the dry blend is shown in Table 2.

2.3 GROUT SPECIMENS

The grout slurry was prepared at the 222-S Laboratory using a solids-to-liquid ratio of 8.40 lb of dry mix for each gallon of simulated liquid waste, as indicated in Table 2. See also, Powell, 1994. The simulated liquid was heated to 45 °C prior to mixing. The dry mix, at

ambient temperature, was added to the liquid simulant. Mixing conditions were 30 s on low speed, followed by 30 s on medium speed of the mixer (Lokken et al. 1993).

Table 1. Make-Up of Simulated DSSF Liquid Waste.

Compound	Amount g/L	Compound	Amount g/L
NaNO_3	8.58	$\text{Na}_2\text{B}_4\text{O}_7$	0.131
$\text{NaOH}(50\%)$	149.7	Na_2SO_4	3.89
$\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$	128.0	$\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$	0.302
$\text{Na}_3(\text{PO}_4) \cdot 12\text{H}_2\text{O}$	74.4	$\text{Ca}(\text{NO}_3)_2 \cdot 4\text{H}_2\text{O}$	0.438
NaNO_2	36.9	$\text{Na}_4(\text{EDTA}) \cdot 2\text{H}_2\text{O}$	1.415
Na_2CO_3	36.2	$\text{Na}_3(\text{HEDTA})$	5.293
KCl	1.83	Glycolic Acid	0.645
NaCl	2.50	$\text{Mg}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$	0.028
$\text{Na}_3(\text{Citrate}) \cdot 2\text{H}_2\text{O}$	2.50		

The grout specimens were maintained at 100% relative humidity during curing and subsequent storage to retard evaporative liquid loss. The temperature during curing was ramped from 40 to 45°C in the first hour after pouring; then from 45 to 47°C in the next 1.5 h; from 47 to 55°C in the next 2 h; and finally from 55 to 65°C in the next 9 h. The temperature was held at 65°C to complete a total time period of 27 days. At the beginning of the 28th day, the specimens were cooled to the ambient temperature over 24 hours. This thermal programming attempts to simulate the thermal conditions expected in the grout vault for the first 28 days of curing.

The as-fabricated weights of the six specimens are contained in Table 3. The average weight of the 6 specimens was 2445.1 grams with a standard deviation of ± 64.6 grams.

Table 2. Composition of Dry Mix and Grout Mix Ratio.

Component	Amount
Type I/II Portland Cement	20.7 wt%
Class F Fly Ash	68.3 wt%
Attapulgate Clay	11.0 wt%
Solids to Liquid Ratio	8.40 lb/gal

Table 3. Weights of the Simulated Grout Samples

Specimen #	Weight, gm
1	2348.0
2	2428.8
3	2462.6
4	2452.4
5	2430.9
6	2548.0

Subsequent storage conditions for the grout specimens were maintained at ambient room temperature, $25 \pm 2^{\circ}\text{C}$ ($77 \pm 4^{\circ}\text{F}$), and 100% relative humidity at the specimen. Care was taken to assure that the grout did not lose any liquid by evaporation. The diameter and length and weight of each specimen was determined before it was inserted into the outer irradiation capsule.

3.0 SPECIMEN IRRADIATION TESTING

Four of the six prepared grout specimens were irradiated in the PNL Gamma Irradiation Facility in the 3730 Building of the 300 Area. The remaining two specimens were slated to go into an irradiation tube that did not become available for our use and therefore those capsules served as blank experiments to document the chemical gas generation rates of the grout. All specimens were enclosed in an inner stainless steel container with a lid with a 1" diameter hole that was plugged with a rubber stopper during curing. Each container had an insert with a 3.18 mm (1/8 inch) diameter hole in the bottom lid welded into place prior to irradiation. This small hole was for liquid and gas release and to decrease the rate of water vapor flow to the cooler walls of the outer capsule. The specimens were placed inside stainless steel leak-tight capsules as shown schematically in Figure 1.

The capsules included a valved gas sampling port, a thermocouple, a pressure transducer and a cylindrical well at the bottom of the capsule to capture and retain any released liquids. The chromel-alumel thermocouples were internally grounded. Because of the relatively low heat generation rates (less than a watt in each specimen) the gradients in the grout and associated systems were expected to be small. The depth of the liquid in the well was monitored during the irradiation period using an ultrasonic echoing device that was applied when the system was extracted from the gamma pool.

The capsules were suspended and located inside two of the 15.2 cm ID (6.0 in) tubes (Tubes 17 & 18) of the Gamma Irradiation Facility pool by rigid pipes at positions that provide the desired gamma field for each specimen. (These locations will be discussed later.) Figure 2 shows a schematic arrangement in one of the double-capsule irradiation tubes.

Figure 1. Schematic of Irradiation Capsule used to Investigate Advective Liquid Rejection from Grout.

The temperature and the pressure of each capsule were continuously monitored by a data acquisition system. Three samplings of gas specimens and measurement of liquid levels were performed for each capsule, before, during and at the end of the irradiation period. The irradiation period was 82 days. A small amount of liquid was found in one of the bottom reservoirs and was sampled and analyzed. Because of programmatic changes, it was not possible to perform a full post-irradiation characterization of each specimen to determine the quantity and composition of retained gases in the grout. The only post-irradiation analyses were radiographs of each capsule to determine whether any liquid was present, gas samples taken of the gases released to the capsule, and one liquid sample extracted from the liquid reservoir at the bottom of the capsule.

3.1 APPARATUS DESIGN

A schematic illustration of the capsule is shown in Figure 1. The layout of one of the three test trains as it appears in the irradiation tube of the Gamma Facility is in Figure 2.

Appendix A contains the "Red-Lined" Procedures document for the test. The "RedLining" comments were added and initialed during the course of the test and represent the deviations from the written procedures of the document that were deemed appropriate at the time of the deviation.

Major features of the capsules and test train are called out in these two figures and enumerated below. All six of these irradiation capsules were fabricated to meet the following criteria:

- (1) All components were capable of withstanding gamma radiation fields up to 500,000 R/h for the duration of the experiment (exposures to $2E+08$ Rad);
- (2) Maximum experimental duration was expected to be about 2.5 months;
- (3) All metal parts were of stainless steel;

Figure 2. Irradiation Assembly Layout in the Gamma Irradiation Facility.

- (4) The systems were easily capable of withstanding a maximum pressure of 0.1 MPa (1.0 Atm, 14.7 psig);
- (5) Each capsule could easily withstand a maximum temperature of 90 °C (195 °F);
- (6) The irradiation capsules were inserted inside the 15.2 cm (nominal 6.0 inch) ID tubes in the Gamma Irradiation Facility;
- (7) The positioning components (SST pipes) for each capsule train were marked for angular indexing so that the capsules could be rotated periodically in the Gamma Facility tube to minimize self-shielding effects due to the directionality of the incident gamma field;
- (8) All seals in each capsule were made in such a manner that no extraneous gases were introduced (i.e., no elastomer seals were allowed that might degrade in the radiation to yield organically derived gases);
- (9) The capsules were designed so that, after the irradiation, the specimens were retrievable for examination and characterization, in this case the welds were to be machined open (however, the capsules were not opened for specimen retrieval because of programmatic changes);
- (10) A low-leak-rate system was required, with a maximum leak-rate of less than 1E-07 cc-atm/s, with each capsule component being He-leak checked as needed at critical fabrication points to ensure tightness, and a final, assembled leak check being passed (one capsule did develop a leak during the irradiation due to what was thought to be a faulty Swagelok fitting that failed during handling).

It was necessary to ensure that each capsule contained helium gas prior to the irradiation to create as clean a background as possible for the detection of O₂ and N₂ formation during irradiation testing. A slightly positive pressure of helium also served as a leak checking agent after final weld closure. A method of multiple pressurizations followed by blow-downs was devised to obtain a background air content of less than 1% (see Appendix A).

Sampling devices and monitoring instrumentation incorporated into each apparatus included:

- (1) A valved port through which gas samples were taken for analyses (while the apparatus was withdrawn from the gamma field), with the valve being capable of withstanding the radiation field for the required time and remaining leaktight after multiple reclosures;
- (2) A pressure transducer suitable for providing real-time monitoring of the internal pressure changes in each capsule, capable of withstanding the irradiation environment (the transducers were located outside of the high gamma field, connected to the capsule via small-ID, low volume tubing);
- (3) A volume at the bottom of each capsule that caught any accumulation of liquid that originated from the grout, the depth of liquid was monitored using an ultrasonic method and/or a radiograph of the capsule; and
- (4) A thermocouple to monitor the temperature of the interior of the capsule.

An apparatus external to the irradiation capsule was used for the pre-irradiation inert gas purging and internal volume measurements. This device consisted of a manifold with necessary valving and a vacuum pump that was also used during gas sampling. The irradiation

capsules were withdrawn from the gamma field part way through the irradiation period and samples of gas in the container were taken and tests were done to detect any liquid in the well. The gas specimens in collection vials were transferred to the analytical facilities for compositional analyses.

The temperature and pressure instrumentation was monitored by a calibrated HP 3497A data acquisition system (DAS). The procedures for using the DAS and a description of its software is contained in the Data Package (Feigenbutz, 1993). The system was programmable for ranges and sampling period. The ranges were set for the full range output of each type of transducer. The DAS was programmed to convert the voltage inputs to engineering parameters. The temperature measuring devices were Type-K (Chromel-Alumel) thermocouples. The millivoltage to temperature conversion for Type-K TCs was built into the DAS. The pressure transducers output was from 0 to 50 psia for a 0 to 5 volt output. Calibration curves were obtained for the DAS and the pressure transducers. The certified calibration information from the thermocouple manufacturer was used.

The temperature and pressure was recorded hourly during the irradiation test. Each data record of 6 pressures and temperatures, plus the date and time of the reading, was recorded onto an HP-formatted 3.5" floppy disk. This disk was exchanged weekly. The data on the HP-formatted disk was read using a special HP-Basic program on an IBM-compatible PC and rewritten to a PC-formatted floppy disk as an ASCII file. This file was then transferred to the hard disk of the primary analysis PC where it was imported into a Quattro Pro for Windows Version 5 spreadsheet. The data was converted in various way that aided the analysis and several other spreadsheet files were fabricated. All final graphics were done in the QPro program. The final data file containing all of the information and plots was approximately 5 megabytes long.

3.2 Self-Shielding of Gamma Field

Because of geometric considerations and the self-shielding effects of the gamma radiation by the grout specimens, a calculation of the effective gamma intensity for the specimens was carried out by Technology Support Programs. Since the density of the grout specimen is about 66% greater than water, significant attenuation of the gamma field through the specimen occurred. The magnitude of the local gamma field intensity within the block of grout was calculated using the MCNP code (ORNL 1992). This calculation contains some uncertainty because no drawings were available for the Gamma Facility showing the configuration of the source cage and its cobalt-60 elements. In addition, while the current source strength of each pin is fairly well characterized, the complete internal composition, density and the location of each pin in the holder cage is not.

Using photographs of the facility, in combination with available drawings of the source pins and a few direct measurements of tube spacings and dimensions, a best-estimated geometry was developed for the source configuration relative to the locations of the 6 inch ID tubes. A simplifying assumption was made that the total magnitude of the source was evenly distributed in a cylindrical annulus of the proper diameter, thickness and height. The density of the source pins, as an unknown, was varied within reasonable bounds until the calculations for the dose rate in an empty tube closely matched the decay-corrected values provided by Pacific Northwest Laboratories that were based upon gamma intensity measurements made in 1984.

Using this model, the gamma intensity attenuation factors were calculated for Tubes #17 and #18 that described the average dose rate experienced by a grout specimen as a function of location in the tube. The intensity correction factors become smaller as the grout

Appendix E of Hinman, 1993, contains a letter-report by FN Eichner transmitting the results of measurements of the gamma fields as a function of the location above the bottom of each tube for Tubes #16, #17 and #18. Table 4 contains the results of these measurements for Tubes #17 & #18 where units have been converted from R/min to kR/hr. (Tube #16 was not used in the irradiation experiment.)

Table 4. Measured Gamma Intensities in PNL Gamma Facility Tubes.

Position from bottom of tube (in.)	Tube 16 kR/h	Tube 17 kR/h	Tube 18 kR/h
0	174.6	66.2	51.0
6	456.0	96.0	105.6
12	450.0	90.0	98.2
18	256.0	51.0	60.5
24	58.3	18.1	19.5
30	16.6	7.3	8.1
36	6.7	3.1	4.1

These data are plotted in Figure 3 as the individual open points, designated as "Meas". The measured values compare favorably to the expected gamma intensity curves for the facility obtained from PNL when corrected to April 04, 1993, the date of the measurement.

3.3 EXPERIMENT DESIGN

The goal exposure for the synthetic grout specimens is $1.1\text{E}+08$ Rad. This is the expected exposure to which the grout in the grout vault will be subjected (Roblyer 1993). This assumes an in-grout starting gamma field strength

Figure 3. Gamma Intensity Maps for Tubes #16 & #17 in the Gamma Irradiation Facility. Points are shown for both extrapolations from decay curves and recent measurements.

of 310 Rad/hr and a decay rate that is dominated by the ^{137}Cs (half-life 30.2 yr). Figure 3 shows a decay-adjusted distribution of the ^{60}Co (half-life 5.26 yr) gamma intensity in chosen tubes in the PNL Gamma Facility. These values are for April 1993, the date that gamma intensity measurements were completed for these tubes. Table 5 contains the experimental matrix for the test series and includes the locations in the Gamma Test Facility derived from the information shown in Figure 3, but extrapolated along the decay curve from the April 1993 date to the starting irradiation date of August 23, 1993. During the 80 days of irradiation, another 2.9% reduction in gamma field strength occurred due to ^{60}Co decay and is ignored for the purpose of calculating the total doses for the specimens. Original plans called for the insertion of Specimen #'s 3 & 5 into Tube 16. However, this tube was not available to us and these two capsules remained outside of the gamma field, serving as blanks for comparison to the irradiated specimens. It was found that there was measurable gas generation in these unirradiated capsules, as will be discussed later.

Table 5. Test Matrix for Irradiation Conditions of Synthetic Grout Specimens.

Specimen #	Tube #	Dist fr/Bot in.	BareTube DoseRate kR/hr 12/01/92	BareTube DoseRate kR/hr 09/23/93	Self Shield Corr MCNP	Initial Grout Dose Rate kR/hr	Nominal Irrad. Time mo	Total Dose Rad.
2	18	11.0	113	101.6	0.60	60.8	2.63	1.17E+08
1	18	22.0	37	33.2	0.52	17.4	2.63	3.36E+07
4	17	11.0	113	101.6	0.60	60.8	2.63	1.17E+08
6	17	35.0	3.8	3.4	0.35	1.2	2.63	2.30E+06

During the irradiation period, the internal pressures of all the capsules were monitored continuously using the pressure transducer and data acquisition system. Pressures and temperatures were recorded hourly by the data acquisition system, assuring adequate data quantity and quality for trend interpretation.

Because of the self-shielding effects of the grout mass to the gamma radiation, the capsules were rotated 120 degrees weekly to distribute the dose accumulation about the circumference.

The capsule liquid level in the bottom reservoir and composition of the released gases in the capsule were checked part way through the irradiation period. An ultrasonic transducer was used to examine for a liquid presence.

The gases in the capsule were sampled by expanding them into evacuated stainless steel bulbs through the gas sampling valve at the top of each capsule. A pre-irradiation set of gas samples was taken 22 days after the original encapsulation and inert gas purging operation. A set of gas specimens was also taken after 24 days of irradiation. A third set of gas samples was taken for compositional analyses at the end of the experiment. These gas samples were analyzed by PNL personnel in a Finnigan MAT-271 mass spectrometer that was fully calibrated on a monthly basis and checked daily with a nitrogen calibration to correct for instrumentation drift.

The capsules were radiographed prior to the beginning and at the end of the irradiation period to ascertain the quantity of liquid in the bottom of each capsule. The amount of radiation received in obtaining these radiographs was negligible, relative to the total radiation received during the specimen's time in the Gamma Facility.

Information obtained during this experiment was entered into the Laboratory Notebook WHC-N-763.

4.0 RESULTS and DISCUSSION

The following text describes the experimental results and observations made before, during and after the gamma irradiation testing of four simulated-waste-containing grout specimens and two that were not irradiated. Significant amounts of gases were generated by the irradiated specimens. Small amounts of gases were generated by the unirradiated grout specimens. The two specimens subjected to the highest (and equal) gamma fields showed dissimilar gas and liquid release characteristics. One of the specimens released about twice as much gas to the capsule as the other, and the other rejected a small amount of liquid. This liquid was collected and analyzed and was found to contain significant amounts of the original salts that were in the simulated liquid waste (but modified in composition). The reason for this dissimilarity in gas and liquid release is not clear. Since the amount of retained gas in the grout specimens after the irradiation could not be determined, it is not possible to know whether the difference in behavior lies in the gas generation rate or in the release rate from the grout matrix. The sections below present and discuss these and other findings from this test series.

4.1 Pre-Test Measurements

When the 28-day cured grout specimens were received in the capsule assembly area where they were to be loaded and welded into the hardware, it was noted that there was some liquid present in two of the stainless steel beakers containing the grout. These were Capsules #3 and #5. It was decided that the originally designated locations for these specimens in the Gamma Irradiation Facility would be changed to lower priority ones because of the apparent non-standard oversaturated condition of the grout (the final locations are reflected in Table 4). Specimen #3 was moved from the top position of Tube #17 of the Gamma Facility to the bottom location in Tube #16. Specimen #5 was moved from the lower location in Tube #16 to the top position of the same tube. At this point in the timing of the preparations for the grout irradiation test, it was becoming apparent that there was a slim probability of getting any specimen into the Tube #16 because of conflicting experiments of a higher priority. Tube #16 was the highest gamma intensity tube in the facility that was large enough to receive these irradiation capsules.

4.1.1 Capsule Purging

It was advantageous to obtain as pure a helium atmosphere within each capsule as possible, because it was expected that one of the gases that may be formed from the radiolytic and/or chemical processes would be nitrogen. It was also of interest to see if any oxygen gas might be formed. Therefore, a process of repeated pressurizations and blow-downs was undertaken for each capsule to assure that the initial gas composition contained only a small amount of residual air. This was done in this manner to maintain a positive pressure on the specimen relative to atmospheric at all times to retard liquid loss by evaporation.

All of the air-containing capsules were cycled between 1 and about 3.4 atm (15 to 50 psia) ten times using 99.99% pure helium gas on July 28, 1993.. The capsules were allowed to remain at the high pressure for a minimum of 10 minutes for each cycle to allow for mixing and interdiffusion of the He and the residual air components before being returned to ambient conditions. It was calculated that this treatment, in theory, should result in an atmosphere containing on the order of 0.01% residual air. Care was taken to maintain a few psi of positive pressure after each blow-down. On the final blow-down, each capsule was left at a positive pressure of about 5 psi above atmospheric. The capsules began their irradiation period in this

slightly pressurized condition. As discussed in the next section, the gas in each capsule was sampled after a 22 day delay.

4.1.2 Gas Sampling

Due to scheduling conflicts at the Gamma Irradiation Facility, gas samples were not acquired from the capsules until 22 days after the internal atmosphere purification steps, 3 days prior to the start of the irradiation period. These samples were analyzed by PNL using the mass spectrometer. The results from these analyses are contained in Table 6. The values in this table indicate the importance of recognizing the 22 day delay between the purging of the capsules and the sampling of the gases. Because of the very small amount of oxygen found in the system, as indicated in Table 6 (0.003 mole%), it was surmised that the purging process was successful in providing a very pure starting atmosphere within the capsules. However, it is apparent from these data that some of the gas (the N_2) either diffused out of the grout after being entrained during mixing and pouring, or some gases were generated in the grout and released to the capsules over the 22 day period (N_2O and N_2). Because of the lack of O_2 and the presence of N_2O , the latter is more probable. Table 7 contains the calculated amount of gases when the He component is computationally removed by subtraction. This subject will be revisited below for two capsules that were not irradiated, but were monitored over the period of irradiation of the remaining four specimens.

**Table 6. Analyses of All Gases from Grout Irradiation Capsules
prior to Irradiation, 22 Days after Fabrication.
Sampled on August 19, 1993.**

Gas Species	Capsule 1 mole%	Capsule 2 mole%	Capsule 3 mole%	Capsule 4 mole%	Capsule 5 mole%	Capsule 6 mole%	Ave mole%	StdDev
Ar	0.026	0.02	0.007	0.007	0.005	0.037	0.017	0.013
CO ₂	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	NA	NA
CO	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	NA	NA
He	96.85	96.83	97.81	98.13	98.34	94.63	97.10	1.37
H ₂	0.0076	0.0118	0.0124	0.0079	0.0108	0.0076	0.0097	0.0022
CH ₄	< 0.001	< 0.001	< 0.001	< 0.001	< 0.001	< 0.001	NA	NA
N ₂	1.74	1.33	0.411	0.397	0.291	2.59	1.13	0.93
O ₂	0.002	0.007	0.002	0.001	0.001	0.006	0.003	0.003
N ₂ O	1.37	1.80	1.76	1.46	1.35	2.73	1.74	0.52
NO _x	< 0.001	< 0.001	< 0.001	< 0.001	< 0.001	< 0.001	NA	NA
Totals (w/o He)	3.17	3.19	2.21	1.88	1.68	5.39	2.92	1.37

4.1.3 Free Volume Calibrations

After the final assembly and internal atmosphere cleanup of each capsule, their free volumes were measured using a calibrated standard volume and accurately calibrated pressure gauges on July 29, 1993. Details of these volume measurements are contained in the Laboratory Notebook, WHC-N-763. An important criterion for any pressure manipulations of the capsules was that the pressure always remain at or above one nominal atmosphere to prevent or minimize evaporative or hydraulic losses of liquid from the grout. For the volume calibrations, a standard volume (523.4 cc) was initially pressurized to about 3 atmospheres (44 psia). This pressure was then vented into a capsule which was initially near ambient atmospheric pressure, and the final, stabilized pressure of the combined volumes was measured using the attached calibrated pressure transducer. An additional check for each capsule was performed where the procedure was reversed, i.e., the standard volume was evacuated to a pressure of less than 20 microns Hg (essentially 0 atm). Then the capsule, that was had been pressurized to about 3 atm, was expanded back into the standard volume and the pressure change again was measured after it had stabilized. The final pressure of the combined volumes (capsule + standard volume) after this step was about 1.2 atm (18 psia). Both of these procedures were carried out twice for each capsule (a total of 4 deltaP's were determined for each capsule). Some hysteresis was noted in comparing the computed volumes for the forward and reverse pressurization procedures. The standard deviation for the four measurements ranged from 0.19% to a maximum of 1.3% of a total volume in the range of 800 cc. The results of these volume measurements are shown in Table 8.

**Table 7. Analyses of Gases from Grout Irradiation Capsules
prior to Irradiation with Helium Computationally Subtracted
August 19 sampling**

Gas Species	Capsule 1 mole %	Capsule 2 mole %	Capsule 3 mole %	Capsule 4 mole %	Capsule 5 mole %	Capsule 6 mole %	Ave	StdDev
Ar	0.82	0.63	0.32	0.37	0.30	0.69	0.52	0.22
CO ₂	< 0.32	< 0.31	< 0.45	< 0.53	< 0.60	< 0.19	NA	NA
CO	< 0.32	< 0.31	< 0.45	< 0.53	< 0.60	< 0.19	NA	NA
He	NA	NA	NA	NA	NA	NA	NA	NA
H ₂	0.24	0.37	0.56	0.42	0.64	0.14	0.40	0.19
CH ₄	< 0.03	< 0.03	< 0.05	< 0.05	< 0.06	< 0.02	NA	NA
N ₂	54.9	41.7	18.6	21.0	17.32	48.0	33.6	16.6
O ₂	0.06	0.22	0.09	0.05	0.06	0.11	0.10	0.06
N ₂ O	43.3	56.4	79.5	77.8	80.4	50.6	64.7	16.5
NO _x	< 0.03	< 0.03	< 0.05	< 0.05	< 0.06	< 0.02	NA	NA
Totals (w/o He)	100.00	100.00	100.00	100.00	100.00	100.00	2.92	1.37

Table 8. Results from Capsule Free Volume Calibration Measurements, July 29 Operation

Capsule #	Free Volume (cc)	StdDev (cc)	SD (%)
1	854.9	7.4	0.87
2	833.5	1.6	0.19
3	824.8	2.5	0.30
4	832.7	11.2	1.34
5	843.2	8.3	0.98
6	735.8	4.0	0.54

4.2 Unirradiated Grout

The two capsules that were not irradiated (Capsules #3 and #5) experienced the pressure fluctuations versus time shown in Figure 4. These pressure histories were dominated by two processes: two gas sampling events and the ambient temperature fluctuations within the Gamma Irradiation Facility building. The displacement between the two curves represents the difference in starting pressure between the two capsules.

Figure 4. Pressure vs time for Two Unirradiated Synthetic Grout Specimens.

The temperature histories shown in Figure 5 occurred because of the lack of heating in the building during off-hours and weekends. Over the course of the test, the two unirradiated capsules were allowed to sit next to the pool where the other four capsules were being irradiated. The temperature in the capsules outside the pool was warmer than those inside the pool and changed diurnally and with weather trends. The water in the pool was kept at a low temperature with a refrigeration system.

Figure 5. Temperatures vs Time for the Two Unirradiated Synthetic Grout Specimens.

Some electronic problems were encountered during the first several days of the irradiation period. It was found that the electronic noise in some of the temperature and pressure inputs was due to a ground loop problem. The ground loop was broken and the collection of the data was relatively straightforward for the rest of the test period.

Figure 6 shows the pressure changes (ΔP) experienced by the unirradiated capsules outside of the pool referenced to the starting pressure at the beginning of the irradiation in August 1993. These pressure changes are still strongly influenced by the temperature changes in the facility. The pressure changes in each capsule were near-identical.

Figure 7 shows the changes in the number of micromoles of gas per gram of grout in these two unirradiated capsules as a function of time. The gas sampling events have been factored out and the temperature variations were (partially) corrected for using the perfect gas law:

$$dn / m = V / (R * m) * (P_1 / T_1 - P_o / T_o) * 10^6,$$

where: dn/m = the number of micromoles of released gas per gram of grout,
 m = the mass of the grout in each capsule,

V = the free volume of each capsule,
 R = the gas constant,
 P_1 = the pressure at any time,
 T_1 = the absolute temperature at any time,
 P_o = the initial pressure at which $dn = 0$, and
 T_o = the absolute temperature at which P_o exists.

Figure 6. Relative Pressure Change vs Time for the Two Unirradiated Synthetic Grout Specimens.

These calculations correcting for temperature history were not able to filter out all the minor diurnal pressure changes because of the location of the thermocouple that provided the temperatures shown in Figure 5. The mass of the grout sample produced a time constant for a thermal lag that prevented a complete elimination of the "ripple" effect showing up in the calculated gas quantities.

The amount of gas in each unirradiated capsule increased with time, as noted above for the first 22 days after final assembly. The addition of gases to the starting helium was confirmed in the three analyses of gas samples taken from the capsules on August 19 (22 days after final closure and 4 days prior to start of the irradiation period), September 15 (after about 550 hr of irradiation on the other four capsules) and November 12 (at the end of the irradiation

Figure 7. Quantity of Gases Released from the Two Unirradiated Synthetic Grout Specimens.

period, about 1940 hr). Tables 6 and 9 contains the results of the gas analyses for these three samplings.

It must be emphasized that the curves shown in Figure 7 represent the gas release rates, not the gas generation rates. This is an important point because of the relatively massive nature of the samples. It must be assumed that gases are being generated uniformly throughout the volume of the sample. Since the grout structure is relatively "tight" to the flow of fluids, a gas molecule generated in the interior of the specimen cannot immediately communicate its existence to the pressure transducer that is looking at the pressure of the free volume of the capsule. In Section 1.2 of this report, background information is provided on this phenomenon, where gas release from the grout is discussed relative to diffusion of the gas through the liquid phase and by advective flow of the liquid through the pore structure of the grout. Without a timely determination of the amount of gases retained in the specimens, we can never directly measure the gas generation rate. Pressure change observations yield only gas release quantities. However, at longer times, the gas release rate approaches the gas generation rate when the specimen becomes saturated, and the uncertainty in generation rates is reduced. This will be discussed below with regard to the measured gas releases from the irradiated specimens where the radiolytic generation rates are higher than the chemical generation rates in the unirradiated samples.

The gas accumulation rate seen in Figure 7 for the #3 Capsule was used to estimate a background chemical gas generation rate to be applied later in this report to at least one of the irradiated capsules. A linear regression analysis of these data leads to a chemical gas release rate of $1.02\text{E-}04$ micromoles/gm hr. This generation rate was used below to make a correction to the gas accumulation rate for only the Capsule #6 that was irradiated at the lowest gamma field intensity (1.2 kR/h) because the rate of release of chemical gas is a significant portion of

the total release rate. The correction was not applied to any of the other capsules because the chemical gas addition is assumed to be insignificant at the higher radiolytic gas release rates.

Table 9. Gas Analyses for Gases Released to the Unirradiated Capsules #3 & #5 During the Test Period

Gas Species	Capsule 3				Capsule 5			
	Sept 15 Sampling mole %		Nov 12 Sampling mole %		Sept 15 Sampling mole %		Nov 12 Sampling mole %	
	with He	w/o He	with He	w/o He	with He	w/o He	with He	w/o He
Ar	0.011	0.31	0.18	0.82	0.008	0.28	0.15	0.79
CO ₂	< 0.01	< 0.28	< 0.01	< 0.05	< 0.01	< 0.34	< 0.01	< 0.05
CO	0.028	0.79	< 0.001	< 0.00	0.032	1.10	< 0.01	< 0.05
C ₂ H ₆	< 0.001	< 0.03	< 0.001	< 0.00	< 0.001	< 0.03	< 0.001	< 0.01
He	96.48	NA	78.2	NA	97.13	NA	81.1	NA
H ₂	0.021	0.59	0.032	0.15	0.019	0.65	0.03	0.16
CH ₄	< 0.001	0.03	< 0.001	< 0.00	< 0.001	0.03	< 0.001	< 0.01
N ₂	0.623	17.51	14.3	65.43	0.423	14.56	12.1	63.74
O ₂	0.013	0.37	3.68	16.84	0.01	0.34	3.14	16.54
N ₂ O	2.82	79.26	3.65	16.70	2.38	81.93	3.54	18.65
NO _x	< 0.01	< 0.28	< 0.001	< 0.00	< 0.001	< 0.03	< 0.001	< 0.01
NH ₃ (est)	< 0.01	< 0.28	< 0.001	< 0.00	< 0.01	< 0.34	< 0.001	< 0.01
Totals (w/o He)	3.56	100.00	21.86	100.0	2.91	100.00	18.98	100.00

4.3 Irradiated Grout

Figure 8 shows the linear dose accumulation curves for the three gamma irradiation intensity locations in Tubes #17 and #18. The information contained in Table 4 was used to derive these curves. Capsules #2 and #4 are in equivalent (highest) dose rate locations. Capsule #1 was placed in an intermediate dose rate location and #6 at a very low dose rate (see Table 4).

The accumulated pressure and temperature histories over the test period within each of the four irradiated capsules are shown in Figures 9 and 10. All of the pressure curves show step decrease on September 15 and November 12, a result of the collection of gases for analyses at these times. Capsule #1 developed a leak during the September sampling. The leak was reduced when the test train was re-extracted from the irradiation tube and various fittings were adjusted. It was determined that the leak occurred at a compression fitting on the capsule lid that led up to the pressure transducer through thin-walled capillary tubing. Even after the leak rate was reduced, enough leakage remained that subsequent pressure data from this capsule was not useful.

Figure 8. Accumulated Dose for the Four Gamma-Irradiated Synthetic Grout specimens. #2 refers to capsule, B means bottom location, T is top, 17 is Irradiation Facility Tube number 17, etc.

Figure 9. Pressure vs Time for the Four Gamma-Irradiated Synthetic Grout Specimens.

Figure 10. Temperatures vs Time for the Four Gamma-Irradiated Synthetic Grout Specimens.

The temperature records for the irradiated capsules shown in Figure 10 indicate that the variations in temperature over the irradiation period of from August to November were much smaller than for the unirradiated capsules shown in Figure 5. This is because the irradiated capsules were contained in submerged tubes in the Gamma Irradiation Facility's water pool. The water is used as shielding against the gamma field. Because of the thermal inertia of the mass of water in the pool, and the fact that it was chilled with a cooling system, the temperatures of these capsules were maintained at a far more constant temperature than existed outside the pool that changed with the day-night and Fall into Winter progression of ambient conditions. The measured pool temperature varied between 16.7°C (62°F) and 17.2°C (63°F) throughout the irradiation period. All of the measured temperatures were at or greater than these temperature.

The two capsules placed in the highest gamma intensities, #2 and #4 (both at 61 kR/h), were at the highest temperatures, being about 1.4°C warmer than the measured pool temperature. These two capsules operated within a few tenths of a degree C of each other, indicating that they were in very similar gamma fields (this will be discussed in more detail later in this report). In a similar manner, the temperatures of Capsules #1 and #6 (17 and 1.2 kR/h, respectively) fell into line relative to the gamma field strengths to which they were subjected.

The temperature increase and subsequent variations after November 13 reflect the ambient conditions in the Gamma Irradiation Facility building and track the same temperatures for that period as shown in Figure 5 for the unirradiated Capsules #3 and #5.

Figure 11 shows the pressure changes (ΔP) in the four irradiated capsules, adjusted to the beginning pressure that occurred after the initial cool-down of the capsules upon insertion into the Gamma Facility pool. The pressure decreases due to the gas sampling events are still evident. These curves differ from those of Figure 9 in that the vertical scale in Figure 9

represents the absolute pressure in each capsule as the irradiation period progressed, and for several weeks after being removed from the gamma field. Figure 11 is plotted as the pressure gain from an arbitrarily chosen zero-point. The pressures shown in Figure 11 are directly related to the accumulation of gases in the capsule free volume, in addition to the thermal

Figure 11. Relative Changes in Pressure vs Time for the Four Gamma-Irradiated Synthetic Grout Specimens.

effects of the temperature history shown in Figure 10.

When the pressure changes due to the factors mention in the previous paragraph (gas sampling and temperature changes) are factored out (see the discussion of the computational methods in Section 4.2 above), the resulting gas release curves shown in Figure 12 are obtained. These points that make up these curves were computed using a form of the perfect gas law to compute the number of moles of gas that were released from the grout to the free volume of each capsule.

The gas release and dose rate data was used to calculate an effective G-Factor for total gas generation for all four of the irradiated capsules. The formula for this calculation is as follows:

$$G_g = dn * N / (Dose * E_g * Mass)$$

where:

- G_g = the effective G-factor for total radiolytic gas generation, in molecules/100 eV,
- dn = number of moles of gas released to the capsule,
- N = Avogadro's Number, $6.023E+23$ molecules/mole,
- $Dose$ = the total dose of the grout specimens, Rad,
- E_g = $6.24E+11$ (100 eV)/(gram*Rad) deposited energy conversion factor, and
- $Mass$ = the number of grams of grout (see Table 3).

The description of G_g as being an effective G-factor is made for the same reasons as the discussion above for the difference between generation rate and release rate. G_g is determined using the released gases that can be detected by the pressure transducers and not the true gas generation rate. Since the grout sample should be acting as a capacitor for gases, there should be a point at long times where the system reaches a steady-state where the release rate equals or asymptotically approaches the generation rate. The calculated G_g functions are plotted in Figure 13. It can be seen that these curves act in the way described, where it appears that the G-factor for total detected gas tends to reach a constant value, indicative of a possible steady-state condition.

Figure 12. Quantity of Gases Released from Specimens of Gamma-Irradiated Synthetic Grout.

Because of the relatively slow gas generation rate in the lowest gamma field capsule, the G_g determined from Capsule #6 includes a correction for the chemical gas generation observed in the unirradiated capsules, as discussed in Section 4.2 and shown in Figure 7 above. The data making up this curve has had a linearly determined, temperature sensitive, amount of gas subtracted from the dn values, based upon the unirradiated gas buildup in Capsule #3. Table 10 shows the methodology for the temperature corrections for the chemical gas generation rates. The activation enthalpy for the chemical gas generation rate was taken from SY-101 tank gas generation rate studies (Goheen, et al., 1993). At the relatively low irradiation field (still about 4 times greater than the maximum computed for the grout vault), the chemical generation rate calculated from the unirradiated capsule makes up more than 30% of the total observed gases released to the free volume of Capsule #6. No correction was made to rates for Capsules #2 or #4 because of their higher relative rates.

Figure 13. Effective G-Factors for Radiolytic Gas Formation in the Four Gamma-Irradiated Synthetic Grout Specimens.

Table 10. Methodology for Temperature Correction to Chemical Rates

Curve fit of data in Figure 7 yields:

Slope: $1.024\text{E-}04 \text{ umole}/(\text{gm hr})$

Intercept: $0.019 \text{ umole}/\text{gm}$

Gas Generation Rate Adjustment:

Ave. T f/Unirradiated Capsules: $21.04 \text{ }^{\circ}\text{C} = T_{\text{ave}}$

R (gas const): $1.9872 \text{ cal}/(\text{deg mole})$

ΔH (activation enthalpy): $21,500 \text{ cal/mole (f/SY-101 Studies)}$

$$\text{Arrhenius RateAdjFactor} = \exp((\Delta H/R) * ((1/(T_{\text{avg}} + 273)) - (1/(T_{\text{irr}} + 273))))$$

Capsule #	1	2	4	6	
T_{irr} :	17.4	18.4	18.4	17.0	$^{\circ}\text{C}$
RateAdjFactor:	0.628	0.715	0.719	0.597	dimensionless
AdjRate:	$6.42\text{E-}05$	$7.32\text{E-}05$	$7.36\text{E-}05$	$6.11\text{E-}05$	$\text{umole}/(\text{gm hr})$
TotReleaseRate:	Leak	$7.02\text{E-}03$	$3.44\text{E-}03$	$2.39\text{E-}04$	From Fig. 12
TotRel/AdjRate:	N/A	96	47	3.9	

4.4. Post-Irradiation Observations

The composition of the gases from the two unirradiated capsules has already been presented in Section 4.2 above.

At the completion of the irradiation test period, the four capsules that had been in the gamma fields were allowed to remain undisturbed for 17 days to investigate the gas release/generation behavior after termination of the irradiation. This period of operation can be seen in Figures 9 through 12 above. No gas sampling was done at the end of this period. There seems to be a reducing rate of gas release to the capsule, as might be expected if the gas generation rate was reduced or went to zero upon removal from the gamma field. In this case, the grout mass acts like a capacitor for the temporary storage of gas being released through the resistance of the grout structure. The specimens experienced an increase in temperature from that experienced in the Gamma Facility Pool. The temperature in the pool was on the order of 17 to 18°C and after being extracted from the pool, the temperature went to 18 to 22°C, as documented in Figure 10. The higher out-of-pool temperature may have affected the chemical generation rates in the specimens.

The capsules were all radiographed to determine if any liquid was present in the collection cup at the bottom of the capsules. It was found that liquid was present in only one of the capsules, Capsule #4, that had experienced the highest dose rate (along with Capsule #2) of 61 kRad/hr to a total dose of 1.2E+08 Rad. This liquid was collected for analysis by drilling a small hole in the bottom of the cup and pouring it out into a collection vial. The hole in the capsule was subsequently rewelded closed. The resultant liquid was bright yellow in appearance. The liquid was determined to weigh 8.60 grams. The analysis of the liquid found that the specific gravity was 1.0753 grams/cc (see Table 13). The computed volume of the liquid is 8.00 cc.

The results from the analyses of this liquid for selected ions are shown in Table 13. Also shown in this table are the starting concentrations for the components as analyzed in the initially prepared liquid waste simulant (Powell, 1994). It can be seen that the concentrations have changed significantly, usually decreasing from that of the beginning simulant. An exception to that observation is the chromium ion. It appears that there was some dissolution of the stainless steel by the liquid solution in the capture cup (an analysis for other components of the stainless steel was not done). A notable missing component in the collected liquid is the hydroxide ion, which appears to have been drastically reduced as indicated by the measured pH of 9.48, which is only to the slightly basic side of neutral (pH 7). It is surmised that the missing hydroxide was retained in the grout because there is no recognizable way that components of the system, including the stainless steel or any contaminants thereof that could neutralize the hydroxide. Another chemical component that has been depleted from the liquid that was collected was the phosphate ion. This ion was also most likely retained within the cementitious matrix by chemical bonding in the grout because the phosphate ion is a recognized cement-forming component (Kingery, 1976).

**Table 11. Capsule Gas Compositions
for the September 15 Sampling**

Gas Species	Capsule 1 mole %		Capsule 2 mole %		Capsule 4 mole %		Capsule 6 mole %	
	with He	w/o He	with He	w/o He	with He	w/o He	with He	w/o He
Ar	0.035	0.33	0.027	0.14	0.01	0.10	0.041	0.58
CO ₂	< 0.01	< 0.10	< 0.01	< 0.05	< 0.01	< 0.10	< 0.01	< 0.14
CO	0.11	1.05	0.31	1.66	0.11	1.12	0.066	0.94
C ₂ H ₆	< 0.001	< 0.01	< 0.001	< 0.01	< 0.001	< 0.01	< 0.001	< 0.01
He	89.5	NA	81.4	NA	90.25	NA	93.00	NA
H ₂	1.84	17.50	3.9	20.89	2.096	21.43	0.254	3.62
CH ₄	0.001	0.01	0.002	0.01	0.001	0.01	0.001	0.01
N ₂	5.78	54.98	10.4	55.70	4.2	42.95	3.08	43.84
O ₂	0.414	3.94	0.61	3.27	0.35	3.58	0.012	0.17
N ₂ O	2.3	21.88	3.39	18.16	2.98	30.47	3.54	50.38
NO _x	< 0.001	< 0.01	< 0.001	< 0.01	< 0.001	< 0.01	< 0.001	< 0.01
Other HCs	< 0.01	< 0.10	< 0.01	< 0.05	< 0.01	< 0.10	< 0.01	< 0.14
NH ₃	< 0.01	< 0.10	< 0.01	< 0.05	< 0.01	< 0.10	< 0.01	< 0.14
Totals (w/o He)	10.51	100.00	18.67	100.00	9.78	100.00	7.03	100.00

A possible mechanism for the existence of liquids in the cup at the bottom of each capsule is evaporation/condensation from the moist grout specimen to the cooler walls of the capsule. In this process, water is evaporated from the surface of the saturated grout specimen and transported as a vapor to the cooler walls of the capsule where it is condensed as pure liquid water. This water would run down the sides of the capsule and be collected in the cup. Figure 10 indicates that the largest temperature difference measured by the thermocouple in contact with the grout specimen and the pool water occurred in Capsules #2 and #4. This difference is on the order of 1.5°C. The temperature of the capsule wall is greater than pool water, but less than the grout. This is because the heat flows from the grout to the pool which is a heat sink. In addition, the surface area of the moist grout is reduced because the outer skin of the stainless steel beaker covered it. Also, the lid with the 0.125 inch diameter hole further restricted the flow or diffusion of the moisture to the outer walls. Therefore, the driving force for the transport of water vapor from the grout to the cooler metal wall was relatively low.

**Table 12. Capsules Gas Compositions
for the November 12 Sampling**

Gas Specie	Capsule 1 mole %		Capsule 2 mole %		Capsule 4 mole %		Capsule 5 mole %	
	with He	w/o He	with He	w/o He	with He	w/o He	with He	w/o He
Ar	0.16	0.037	0.067	0.12	0.18	0.40	0.2	0.79
CO ₂	< 0.01	< 0.02	< 0.01	< 0.02	< 0.01	< 0.02	< 0.01	< 0.04
CO	0.98	2.25	0.86	1.58	0.18	0.40	< 0.01	< 0.04
C ₂ H ₆	< 0.001	< 0.00	< 0.001	< 0.00	< 0.001	< 0.00	< 0.001	< 0.00
He	56.5	NA	46.4	NA	54.4	NA	74.6	NA
H ₂	5.24	12.04	10.2	18.99	5.88	12.90	0.76	2.99
CH ₄	< 0.001	< 0.00	< 0.00	< 0.00	< 0.001	< 0.00	< 0.001	< 0.00
N ₂	28.6	65.71	34.8	64.75	30.5	66.94	17.1	67.31
O ₂	4.5	10.34	2.64	4.90	4.88	10.71	3.65	14.37
N ₂ O	4.03	9.26	5.16	9.64	3.93	8.63	3.67	14.45
NO _x	< 0.001	< 0.00	< 0.001	< 0.00	< 0.001	< 0.00	< 0.001	< 0.00
NH ₃ (est)	< 0.001	< 0.00	< 0.001	< 0.00	< 0.001	< 0.00	< 0.001	< 0.00
Totals (w/o He)	43.52	100.00	53.65	100.00	45.56	100.00	25.40	100.00

The transport of water vapor from grout to container wall had been postulated in a previous experiment (Lokken, 1993) to explain the presence of liquid at the bottom of the capsule that contained simulated grout specimens with free surfaces. This liquid was relatively dilute in the salts that made up the liquid in the grout. It was because of this observation that our experiment utilized almost totally contained grout specimens. Each grout specimen was initially poured into a 2 liter stainless steel flask that had a lid welded onto it. See Section 3.0 of this report and Feigenbutz, 1993, for a more detailed description of the hardware. It is believed that these factors, small temperature difference and mostly contained specimen, reduced, or even eliminated the transport of water vapor in this experiment.

The most compelling observation that water vapor transport in a thermal gradient was not the source of or did not cause any dilution of the liquid specimen in Capsule #4 is that there was no liquid present in Capsule #2, as seen in the post-irradiation radiographs of the capsules. These two capsules experienced the same environment during the irradiation test. However, there is some basic difference between these two capsules, in that the gas release characteristics of the two specimens were a factor of 2 apart.

Table 13. Measured properties of the Liquid collected from Capsule #4.

Specie	Measured Concentration mg/L	MolWt	Beginning Mole/L*	Ending Mole/L	Change
Cr⁺⁺⁺	9,200	51.996	0.0098	0.177	+1,700%
NO₂⁻	19,100	46.006	0.5360	0.415	-22.6%
NO₃⁻	44,300	62.005	0.9747	0.714	-26.8%
PO₄⁻⁻⁻	< 500	71.741	0.1028	< 0.007	-93.2%
OH⁻	NA (pH<10)	17.000	0.4247	NA	NA
Total Carbon	1,880				
Total Organic Carbon	1,590				
pH = 9.48		SpecGrav = 1.0753 gm/cc		Mass = 8.60 gm	

* Powell, 1994, Table 1.

The reason for the difference in gas release between the two highest exposure (rate) specimens is not clear. The observation that they were both maintained at the same temperature during the irradiation period indicates that the power deposition was the same for each. It is not likely that the experiment in Tube #16 could have absorbed enough radiation to make a factor of two difference in the gamma dose rate in Tubes #17 and #18. The results of the gas analyses shown in the bottom row of Table 11 show roughly twice as much gas present in Capsule #2 (18.7 mole%) as in Capsule #4 (9.8 mole%), indicating that the pressure transducers for both capsules were both indicating reasonable results. Table 12, however, shows an end-of-radiation (Nov, 12) ratio of gas quantities of 1.2 (53.7/45.6) between Capsules #2 and #4, whereas the ratio of the pressure changes prior to the gas sampling on Nov.12, from Figure 11, indicates a ratio of about 2.1 (13.2/6.3).

It is felt that the gas release data is sound, i.e., more gas was released by the grout specimen in Capsule #2 than in Capsule #4. It is further felt that the total dose for both of these two capsules are very near to being equal. Therefore, the most probable thing that could be different between the two specimens is something in their basic makeup. Unfortunately, there is no way of knowing because it was not possible to trace individual specimens through the batching operations during their preparation. It was also not possible to do any post-irradiation characterization of the two specimens. But, because of the observations discussed in Section 4.1 on the presence of a small amount of liquid in Capsules #3 and #5, there may have been some variation in the fabrication procedures from batch to batch.

Since variations in processing during the batching, mixing, pouring, and curing of each specimen are not identifiable, one can only speculate about the reasons for the significantly different behavior between the grout specimens in Capsules #2 and #4. If it is assumed that the difference between the two is because of differences in gas release rates, not gas generation rates, then there should be significantly more gas retained in the #4 specimen. Since this will

not be measured (in the original plan, altered by sudden funding constraints [cancelled program] this measurement would have been done), this explanation will go undetermined. Additional gas retention in #4 would go a long way in explaining the appearance of the advectively rejected liquid. More gas retention leads to higher pressures within the grout matrix. An examination of the G-factor curves in Figure 13, discussed below, shows that the G-factor for Capsule #2 appears to have reached a steady-state condition. The G-factor curve for Capsule #4, however, still appears to have a finite slope to it, indication non-steady-state conditions, commensurate with the condition where the release rate has not yet caught up with the generation rate.

If the gas release/liquid advection mechanism is indeed this sensitive to small variations in processing variables during grout preparation, then it would be advantageous to stay on the high gas release side of that particular variable (e.g. liquid or clay content) to promote a less severe internal pressurization of the grout mass and the possibility of liquid return.

No liquid was observable in any of the other irradiated capsules. Therefore, it is felt that all of the liquid collected from Capsule #4 was placed there by the advective rejection of the chemically modified liquid phase in the grout by internal pressurization due to radiolytically formed gases. The relative amount of liquid rejected by the specimen was small, on the order of less than 0.5 wt%.

5.0 CONCLUSIONS

It was found that, for the specimen geometries used in this test series, and for peak radiation dose accumulation rates on the order of 4 to 60 times of the initial rate expected in the grout vaults (300 Rad/hr), no liquid rejection was observed from 2% to 35% of the target exposure expected in the grout vaults ($1\text{E}+08$ Rad). When the irradiation rate exceeded the projected grout vault dose rate by a factor of 200 a small amount of liquid rejection was observed from one of two specimens that had received 20% more than the goal exposure. Because of other experimental observations involving these two test specimens (gas release rates, gas and liquid compositions, etc.), it was speculated that the difference in behavior between these two seemingly identical specimens was due to unidentifiable specimen preparation dissimilarities.

Because of the differences in the magnitudes of the relative radiation field strengths between this study and an actual grout vault, it is concluded that the potential for liquid rejection by internal gas pressurization from presently configured grout waste forms is very low for the expected conditions. However, because of the great difference in the volume, mass and surface area between the experimental and actual cases, a more sound judgement about the probability of the occurrence of advective rejection of grout liquid awaits a thorough evaluation using the modeling codes.

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APPENDIX A

RED-LINED COPY USED DURING TEST

Test Procedure No. 306E-GROUT, Revision 0
Title: Cobalt-60 Grout Irradiation Test Procedure
Prepared by: L.V. Feigenbutz

Page 1

1.0 PURPOSE

The purpose of this Test Procedure, No. 306E-GROUT, is to provide instructions for performing a gamma irradiation test on six simulated grout specimens in the 3730 Gamma Irradiation Facility.

The purpose of this test is to determine whether liquid is advectively ejected from the grout specimen during gamma irradiation. The secondary purpose is to determine what and how much gases are given off by the grout during gamma irradiation.

To meet these objectives during the test, periodic measurements will be taken to determine the quantity of liquid collected in the chamber well at the bottom. Also, gas samples will be periodically taken and analyzed to determine the composition and quantities of released gases.

2.0 GENERAL DESCRIPTION OF TEST

Six simulated grout specimens, each enclosed in a sealed stainless steel Grout Test Chamber, are to be irradiated in the PNL gamma irradiation facility for a period specified by Grout Technology. As shown in Figures 1, 2, and 3, the six Grout Test Chambers are inserted in gamma irradiation Tubes 16, 17 and 18 as pairs. The test equipment is designed to expose each grout specimen to a specific gamma irradiation dose.

Each chamber is provided with one pressure transducer, one thermocouple and one gas sampling valve. The gas pressure and grout temperature within the six Grout Test Chambers will be continuously monitored and recorded by a data acquisition system (DAS). The Grout Test Chambers will be periodically removed for short times from the irradiation tubes to take gas samples and to measure the liquid level in the bottom well of each chamber by using an ultrasonic probe placed below each chamber well.

3.0 RELATED DOCUMENTS

- 3.1 WHC-SD-ER-TP-005, Rev. 0, July 14, 1993, TEST PLAN FOR GAMMA IRRADIATION OF SIMULATED GROUT SPECIMENS FOR GAS GENERATION/LIQUID ADVECTION TESTS.

4.0 DEFINITIONS

The following terms are defined herein in order to avoid confusion:

- 4.1 **GROUT CASTING CONTAINER or "container"**: The 2 liter stainless steel container which is filled with the synthetic grout mixture. There are six Grout Casting Containers, one for each Grout Test Chamber. The Grout Casting Containers is referred to as "containers".
- 4.2 **GROUT TEST CHAMBER or "chamber"**: The outer, stainless steel chamber which has one Grout Casting Container sealed inside it. There are six Grout Test Chambers. The instrumentation and gas sampling valve is attached to the top cover of each Grout Test Chamber. A "well" is located at the bottom of each chamber, its purpose is to collect liquid ejected from the grout specimen. The Grout Test Chambers is referred to as "chambers".
- 4.3 **GAMMA IRRADIATION TUBES or "tubes"**: The three 16 foot long tubes in the cobalt-60 water pool in the 3730 Building. The chambers will be inserted into these to expose the six grout specimens to various gamma dose rates. The gamma irradiation tubes are referred to as "tubes".
- 4.4 **SUPPORT TUBES**: The 1.25 inch diameter tubes that hold a pair of chambers together, and allow the chambers to be inserted into and removed from, the gamma irradiation tubes.
- 4.5 **GAS CYLINDERS**: Refers to the six stainless steel gas sampling containers that are 25 cc in volume. Also, it refers to the 523.4 cc calibrated stainless steel gas container which is to be used to remove excess gas from the Grout Test Chambers.

5.0 FACILITIES

The grout specimens will be irradiated in the PNL Gamma Irradiation Facility, 3730 Building in the 300 Area. Three gamma irradiation tubes number 16, 17 and 18 will be used to irradiate the six grout specimens. Two grout specimens will be irradiated in each tube at the same time.

6.0 TEST EQUIPMENT

- 6.1 Figures 1, 2 and 3, show the setup in the irradiation tubes for each pair of grout specimens in tubes 16, 17, and 18. The hardware in these figures is described in sketches FE&L-GRT-001 AND FE&L-GRT-002.

The six Grout Test Chambers are numbered 1 thru 6. Each Grout Test Chamber contains an identically numbered Grout Casting Container that contains synthetic grout material supplied by Grout Technology.

6.2 Test Equipment outside Gamma Irradiation Tubes

1. Six 25 cc gas sampling cylinders numbered 1 thru 6 with attached valve.
2. Welch Duo-seal Vacuum pump, Model 1405, for evacuating gas sampling cylinders and manifold prior to taking a gas sample. Hasting Vacuum Gauge, Model VT-6, range 1 micron to 1 atm, for measuring vacuum level in gas sample cylinder and manifold prior to taking gas sample.
3. Ultrasonic examination equipment supplied by WHC NDE.
4. Crane hook Lifting Ring, sketch FE&L-GRT-001, item 23.
5. Installation Support Plate, sketch FE&L-GRT-001, item 25.
6. Welded aluminum stand to support a pair of Grout Test Chambers when gas samples and ultrasonic measurements are taken.

6.3 Pressure Transducers and Signal Conditioning Equipment

Each Grout Test Chamber has an absolute pressure transducer which is connected to the top cover by a 1/16 inch SST tube, located at approximately 8.5 feet above the bottom of the irradiation tubes. The gamma dose at this level is less than 150 rad/hr.

The pressure transducers are Sensotec Model TJE/713-04, 0-50 psia, with minimum accuracy of +/- 0.1% full scale. Operating temperature is -100 to 325°F with temperature compensation from 60 to 160°F. The signal conditioning equipment supplies 10 VDC. The output to the DAS varies between 0 and 5 VDC full scale. The six pressure transducers were calibrated at the WHC Standard Laboratory on 4/8/93. Each calibration is good for one year.

6.4 Thermocouples

Each Grout Test Chamber has a thermocouple (TC) passing through the top cover with the hot junction tip pressed against the top of each Grout Casting Container. Each TC is held and sealed in this position by a 1/16 inch Swagelok fitting welded to the chamber top.

Each (TC) is a Type K Watlow Gordon style AJEA0TA720UK080, 1/16 inch diameter 304SST sheath, 6 feet long, with ungrounded hot junction. The attached lead wire is a type K, fiberglass coated lead, 8 feet long. The lead connector is also located at the 8.5 foot level in each gamma irradiation tube.

The TCs are fabricated to ASTM E207, E220 and E230, and to ANSI MC 96.1, and certification is traceable to NIST (formally NBS).

6.5 Data Acquisition System

One HP 9826 computer.
One HP 3497 data acquisition/control Unit
One HP 91533 hard disk and floppy disk data recorder
One HP ThinkJet printer/plotter

7.0 GENERAL INSTRUCTIONS AND SAFETY

- 7.1 Major deviation from this test procedure and the Test Plan (as specified in Step 3.1) shall be discussed with Grout Technology prior to implementation. Minor modifications may be made, providing they do not affect the intent or purpose of the test.
- 7.2 The instructions in this test procedure may be changed by direction from C.A. Hinman and/or W.J. Powell. All changes shall be entered into this test procedure, using red ink, by L.V. Feigenbutz or his designee. Each occurrence of an approved change is to be initialed and dated.
- 7.3 This Red-Lined Test Procedure and attached Tables and Figures shall be inserted in a 3-ring binder. The 3-ring binder shall remain next to the grout Data Acquisition System in the 3730 Building during the irradiation test.
- 7.4 Persons performing the instructions in this test procedure shall be approved by L.V. Feigenbutz and/or C.A. Hinman. The names of approved persons shall be recorded in Appendix A.
- 7.5 Before the test is started in the 3730 Building, all persons involved in the test shall review this procedure and attend a pre-test review briefing conducted by L.V. Feigenbutz and/or C.A. Hinman. Any person added to the team after this pre-test briefing shall be briefed by L.V. Feigenbutz and/or C.A. Hinman before performing any part of this procedure.
- 7.6 All persons performing the test are required to have a 5-chip dosimeter and a whole body count prior to performing the test in the 3730 Building.
- 7.7 The PNL person in charge of the 3730 Building or his designee, shall be present at all times when WHC personnel enter the control zone around the gamma irradiation pit to perform any test function. *Key issued to WHC personnel to allow them into lab. without PNL being present. WHC persons to work only with DAS when PNL is not present.*
- 7.8 Persons performing the test shall abide by the PNL rules and regulations for the Gamma Irradiation Facility, 3730 Building, and shall follow all PNL procedures for performing operations around the gamma irradiation pit.
- 7.9 All crane lifts in and out of the gamma irradiation tubes shall be conducted by and be under the direction of PNL personnel. PNL shall provide all the safety rules and instructions for these lifts.
- 7.10 All test information not recorded in Tables 1 through 6 and the data acquisition system shall be recorded in the Laboratory Notebook number WHC-N-763, dated and signed.

LVF 8/30/93

PNL Person in charge Monty Teleander allowed WHC personnel to raise & lower tests in tube when there are not 2 PNL persons present.

*MKT
8-30-93*

- 7.11 The test shall not be attended continuously. The times at which:
1) the test should be checked, 2) a data disk changed and gas samples are taken, and 3) liquid levels are measured, shall be specified by Grout Technology, or C.A. Hinman, or L.V. Feigenbutz.

8.0 PRECAUTIONS

CAUTION

- 8.1 After the six Grout Test Chambers are inserted into the gamma irradiation tubes the first time, the six Grout Test Chambers are to remain UPRIGHT at all times until disposition Step 21.0 is performed.

CAUTION

- 8.2 All loose items that may be dropped into the gamma irradiation pool or irradiation tubes shall have a safety line attached. All persons leaning over the pool shall remove loose items from their pockets. Leave the lifting ring, item 23, locked onto the crane hook while over the pool.

9.0 DATA TO BE RECORDED DURING TEST

The following information shall be recorded on the DAS floppy disks, the DAS printer, and in Tables 1 through 6 which are attached to the end of this Test Procedure.

- 9.1 Per DAS operating procedure in Appendix B, record following digital data on floppy disks: date and time, six Grout Test Chamber temperatures and pressures.
- 9.2 NOTE: The Grout Test Chamber temperatures, pressures, time and date, are automatically printed on paper each time this data is recorded on the floppy disks.
- 9.3 Record in Table 1, entitled "DATA ACQUISITION SYSTEM LOG", all changes made to the DAS system, when the changes are made, who made the changes and at whose request.
- 9.4 Record in Table 2, the gamma irradiation pool water temperature.
- 9.5 Record in Table 3, each Grout Test Chamber circumferential position in the gamma irradiation tubes after chamber rotation.
- 9.6 Record in Table 4, the gas sampling data.
- 9.7 Record in Table 5, the ultrasonic measurement data.
- 9.8 Record in Table 6, each chamber pressure and temperature.

10.0 WITNESSES DURING TEST

Anytime a pair of Grout Test Chambers are removed from one gamma irradiation tube, L.V. Feigenbutz and/or Chet Hinman or their designee, shall be present before removal and during reinsertion. These persons do not have to be present when changes are made to the DAS, data is retrieved from the DAS, or when the tests are rotated in the gamma irradiation tubes.

11.0 PRE-INSERTION PROCEDURE INSTRUCTIONS

- 11.1 Mark the top of the three gamma irradiation tubes numbered 16, 17, and 18 at three locations around their perimeter, 120 degrees apart. Mark the locations, CLOCKWISE, as 0°, 120°, and 240°.
- 11.2 Remove the support tubes, items 16, 17, 18, 20, and 21, and place each of the six Grout Test Chambers on the floor next to the gamma irradiation pool and the DAS. Connect the six thermocouples and six pressure transducers to the DAS. Per Step 14.0, operate the DAS and record the six temperatures and pressures per directions given by C.A. Hinman. Step 11.2 shall be continued until C.A. Hinman is satisfied that the test is ready to go into the three gamma irradiation tubes. Before the chambers are inserted into the gamma irradiation tubes, remove the data disk, mark it (in addition to the markings required by Step 14.0) "Pre-insertion Grout Test Data, Disk 0", and give it to C.A. Hinman. Insert a new data disk for recording the initial test irradiation data.
- 11.3 During this pre-insertion evaluation, NDE shall make ultrasonic measurements at the bottom of the six chamber wells. Place a chamber on the aluminum stand to allow the ultrasonic probe to be placed beneath each chamber. NDE shall observe the ultrasonic CRT screen and record the signal pattern as a baseline for future measurements. Record any recordable observations in Table 5.
- 11.4 During this pre-insertion evaluation, take a 25 cc gas sample from each of the six Grout Test Chambers as a baseline. Take the six gas samples to Milt Goheen in the 325 Building for analysis. Refer to Step 17.0 for instructions for taking gas samples. Record gas sample data in Table 4.

12.0 TEST SEQUENCE

HOLD POINT TO PROCEED WITH STEP 12.0

All pre-insertion checkout steps must have been completed to C.A. Hinman's or his designee's satisfaction. The six Grout Test Chambers may be inserted in Gamma Irradiation Tubes 16, 17, and 18, under C.A. Hinman's or his designee's direction.

C.A. Hinman

Date

The test shall be performed in the sequence and at the times described below using procedure Steps 13.0 through 21.0.

- 12.1 Record in controlled Notebook No. WHC-N-763, entitled "COBALT-60 GROUT IRRADIATION TEST", all actions taken, by whom, and when.
- 12.2 Insert the six Grout Test Chambers into the three gamma irradiation tubes using procedure Step 13.0. Step 13.0 is to be used for the initial insertion and all subsequent insertions after taking gas samples and making ultrasonic measurements.
- 12.3 The six Grout Test Chamber's temperature and pressure are to be recorded by the DAS using procedure Step 14.0. Start the recording at the time the chambers are first inserted into the gamma irradiation tubes. The frequency of recording the temperatures and pressures at the start of and during the test, is specified in Step 14.0. Record all pertinent information related to operating the DAS in Table 1.
 1. Every Monday, starting with first Monday after initiating the test, remove the used floppy data disk and put in a new floppy data disk. Give the used floppy data disk to David King.
- 12.4 The DAS will automatically print the time, date, and the six chamber temperatures and pressures every time the time, date, temperatures and pressures are recorded to the floppy disk. This provides a hard copy of the data being recorded to disks.

- 12.5 This Step 12.5 provides the method to periodically look at the test data to determine that the test is progressing satisfactorily. Steps 12.5.1 and 12.5.2 provide the instructions to determine if an action is required when the test data is observed.

Record once every work day in Table 6: the time, date, six chamber temperatures and six chamber pressures. Each entry is to be initialed. Take the temperature and pressure data from the last hard copy print out of the temperature and pressure. If any of the criteria specified below in Steps 12.5.1 or/and 12.5.2 is observed, perform the action required by these steps:

1. When test personnel observe that the pressure in any one of the six Grout Test Chambers exceeds 25 psia, immediately inform either or L.V. Feigenbutz or C.A. Hinman, or their designee. Pressures exceeding 25 psia shall be reduced by Step 19.0 under direction of C.A. Hinman or L.V. Feigenbutz or their designee.
 2. After the test has established a steady-state condition in the rate of pressure rise and in temperature level, if any change in this steady state condition is observed report this change to L.V. Feigenbutz and/or C.A. Hinman.
- 12.6 The gamma irradiation pool temperature shall be recorded in Table 2 at the time the chambers are first inserted. Record the pool temperature once a week after the start of the test. The pool temperature may also be recorded at other times specified by C.A. Hinman and/or L.V. Feigenbutz.
- 12.7 Once a week, the chambers in the three irradiation tubes shall be rotated 120° using procedure Step 15.0. Rotation shall start one week after the chambers are first inserted into the tubes. Record in Table 3 all rotation information.
- 12.8 The six Grout Test Chambers are to be temporarily removed using procedure Step 16.0 each time a gas sample or ultrasonic measurements is to be made.
- 12.9 Gas samples are to be taken from the Grout Test Chambers using procedure Step 17.0. Record gas sample data in Table 4. The times at which gas samples are taken shall be specified by C.A. Hinman and/or L.V. Feigenbutz.

- 12.10 Ultrasonic measurements of the liquid level in each Grout Test Chamber will be performed using procedure Step 18.0. Record ultrasonic measurement data in Table 5. The times at which ultrasonic measurements are to be taken shall be specified by C.A. Hinman and/or L.V. Feigenbutz.
- 12.11 Whenever the pressure in a Grout Test Chamber exceeds 25 psia (approximately 10 psig), the pressure shall be reduced to approximately 15 psia using procedure Step 19.0.
- 12.12 At the completion of the test, the six Grout Test Chambers shall be removed from the gamma irradiation tubes for post-irradiation testing using procedure Step 20.0.
- 12.13 At the completion of the post irradiation testing, the six Grout Test Chambers shall be dispositioned using procedure Step 21.0.

13.0 TEST INSERTION INSTRUCTIONS

Step 13.0 describes the initial insertion and subsequent insertions of the six Grout Test Chambers into Gamma Irradiation Tubes 16, 17, and 18. As shown in Figures 1, 2, and 3, two Grout Test Chambers shall be inserted into each tube.

NOTE: Tube No. 16 may be available only intermittently. The times the pair of chambers are inserted into Tube No. 16 will be specified by C.A. Hinman or his designee.

The location of each of the six Grout Test Chambers in the tubes is specified in the Test Plan, Table 7 (see 3.1). The location of the six Grout Test Chambers are:

<u>TUBE No.</u>	<u>BOTTOM CHAMBER No.</u>	<u>TOP CHAMBER No.</u>	<u>DIST. FROM BOT. OF TUBE</u>
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16	5 3	4 5	11 and 22 inches
17	5 4	6	11 and 35 inches
18	2	1	11 and 22 inches

Changes directed by D&I from Chet Hinman
To L.V. Feigenbutz & Bill Powell, entitled
"modifications to radiation Experiment"
Dated 8/5/93 LVP 8/23/93

- 13.1 Assemble together each set of Grout Test Chambers with support tubes and lock pins as shown in Figures 1, 2, and 3, on the operating deck next to the gamma irradiation pool. **DO NOT ATTACH SUPPORT TUBE ITEM No. 19 AND CONNECTOR ITEM No. 22 AT THIS TIME.** Attach each pair of pressure transducers and thermocouples to the top of support tubes No. 18 and 21. Connect each pressure transducer and thermocouples to the DAS.

HOLD POINT FOR STEP 13.2

EACH SET OF GROUT TEST CHAMBERS ARE ASSEMBLED IN THE ASSIGNED POSITION WITH THE CORRECT SUPPORT TUBES AND ARE READY TO BE INSERTED INTO GAMMA IRRADIATION TUBES. APPROVAL SHALL BE SIGNED BY C.A. HINMAN OR HIS DESIGNEE.

SETUP APPROVED FOR TUBE 16:	_____	DATE:	_____
SETUP APPROVED FOR TUBE 17:	<u>CAH</u>	DATE:	<u>08/23/93</u>
SETUP APPROVED FOR TUBE 18:	<u>CAH</u>	DATE:	<u>08/23/93</u>

- 13.2 The following steps apply to the insertion into any of the 3 gamma irradiation tubes. Refer to Figure 4 for a sketch of the equipment setup and insertion steps.
- 13.3 PNL shall operate the crane.

CAUTION

**VERIFY THAT ALL FIVE LOCKING PINS ARE
INSTALLED CORRECTLY AND THE END OF THE
SAFETY LINES ARE SECURED.**

13.4 Insertion steps:

1. Attach the lifting ring, item 23, to the top of support tube 18 or 21 with a locking pin.
2. Attach the lifting ring to the crane hook.
3. Secure the end of the safety line that is attached to support tube 18 and 21.
4. Carefully lift the pair of Grout Test Chambers up and over the assigned gamma irradiation tube.

5. Slowly lower the chambers down into the tube until the 1/4 inch diameter support rod (welded to top of support tube no. 18 and 21) is just above the top of the gamma irradiation tube.
6. Secure the end of the safety line that is attached to the support plate, item 25. As shown in Figure 4, center the support plate on top of the gamma irradiation tube so that it is securely support by the tube.
7. Slowly lower the chambers until the 1/4 inch diameter support rod is resting on the support plate and is located between the 4 roll pins.
8. Refer to step 3 in Figure 4 for the following. Leaving the lifting ring on the hook, release the ring from support tube 18 or 21. Move the hook and ring to the side of the pool and attach the ring to support tube, item 19. Attach tube connector, item 22, to the other end of the support tube. Move this assembly over the gamma irradiation tube and connect the tube connector, item 22, to the top of support tube 18 or 19.
9. Lift the chambers enough to remove the support plate from the top of the gamma irradiation tube. Slowly lower the chambers into the tube until the bottom support tube, item 16, is resting on the bottom of the gamma irradiation tube.

NOTE: For the initial insertion, rotate the chambers as they are lowered, so that support tube, item 19, is located at 0° as mark on top of the gamma irradiation tube. Record in Table 3 the insertion date and time, and location of the chambers (0°).

10. Remove the lifting ring and crane hook.

14.0 TEMPERATURE AND PRESSURE MEASUREMENTS USING DAS

- 14.1 Each Grout Test Chamber temperature and pressure shall be recorded on data floppy disks by the DAS. The DAS shall be operated using the DAS Operating Procedure in Appendix B.
- 14.2 Record all operations performed on the DAS in the DAS log, Table 1, starting when the test is started as defined by Step 11.3. During the pre-insertion Step 11.0, record in Table 1 any DAS operation information that is requested by C.A. Hinman.

- 14.3 Label all data floppy disks as specified in the DAS operating procedure Step 5.4.3.F when inserted ("IN") into the drive, and Step 5.4.3.D when removed ("OUT") from the drive.

ADDITION LABELING - Write on each label "Grout Disk #___", starting with number 0 for the pre-insertion data recorded in Step 11.0. Record in the DAS log, Table 1, the disk number, date and time the disk is inserted and removed from the drive, and initials.

- 14.4 Data disks that are removed from the DAS drive are to be temporarily stored in the "GDAS USED Data Disks" box while performing work at the 3730 Building. On any given day when work is completed at the 3730 Building, take all used data disks in this temporary storage box to David King who will convert the data to a usable format and record this converted data onto a data disk with the same numbered label as the original, "Grout Disk #___". Deliver the converted data disks to C.A. Hinman.

14.5 INITIAL DATA RECORDING AT START OF TEST IN EACH TUBE

Start recording the temperature and pressure on a data floppy disk and hardcopy print out, starting at the time a pair of chambers are first inserted in a gamma irradiation tube. Continue recording the data every minute for the first 3 hours. Record the initial 3 hours at one minute intervals. This initial recording frequency may be changed by C.A. Hinman or L.V. Feigenbutz.

14.6 DATA RECORDING AFTER THE FIRST 3 HOURS

After the first three hours in the tube, reduce the disk recording frequency and hardcopy print out, to every 10 minutes. This recording frequency may be changed by C.A. Hinman or L.V. Feigenbutz.

- 14.7 Every Monday, starting with first Monday after starting the test, remove the used floppy data disk and put in a new floppy data disk. Give the used floppy data disk to David King.

15.0 TEST ROTATION INSTRUCTIONS

Rotate, **CLOCKWISE**, each pair of Grout Test Chambers in each gamma irradiation tube 120° once every 7 days. Start the rotation for each tube on the seventh day after a test was started in that tube.

The chamber can be rotated counter-clockwise to prevent wrapping of instrumentation leads.

A-14 By C.A. Hinman 9-7-93

- 15.1 Attach the lifting ring and crane hook to the top of the upper support tube, and lift the chambers until the chambers freely rotate.
- 15.2 Rotate the chambers 120° **CLOCKWISE**, and then lower the chambers until they are supported by the bottom of the tube.
- 15.3 Remove the lifting ring and crane.
- 15.4 Record in Table 3 the date/time and initials, and location of chambers in each tube.

16.0 REMOVAL INSTRUCTIONS FOR GAS SAMPLING AND ULTRASONIC MEASUREMENTS

Step 16.0 describes the temporary removal of a pair of Grout Test Chambers from a gamma irradiation tube to take a gas sample from and measure the liquid level in each chamber. The chambers will be set down along side the pool for this work. This Step 16.0 may also be used for final removal when the test is completed.

CAUTION

- 16.1 After the six Grout Test Chambers are inserted into the gamma irradiation tubes the first time, the six Grout Test Chambers are to remain **UPRIGHT** at all times until disposition Step 21.0 is performed.
- 16.2 PNL shall operate the crane.
- 16.3 Removal steps. These steps are the reverse of the steps given in Step 13.4 and are abbreviated here:
 1. Lift the chambers using the lifting ring and crane hook, until the 1/4 inch diameter rod at the top of the lower support tube, item 18 or 21, is just above the top of the gamma irradiation tube.
 2. Place the support plate on top of the tube, and support the chambers by placing the 1/4 inch rod between the 4 roll pins on the support plate.

3. Remove the upper support tube, item 19, and tube connector, item 22, and reconnect the lifting ring to the top of the lower support tube, item 18 or 21.
4. Lift the chambers out of the gamma irradiation tube and move them to the side of the pool. Lower the chambers until the lower chamber is resting on the aluminum support table. Continue to support the column with the crane hook during gas sampling and ultrasonic measurements.
5. Return the chambers to the gamma irradiation tube using Step 12.4 after taking the sample and making the measurements, .

17.0 GAS SAMPLING INSTRUCTIONS

Step 17.0 describes gas sampling of each of the six Grout Test Chambers. The chambers are sitting on the operating deck alongside the gamma irradiation pool for this sampling Step 17.0.

- 17.1 Record all gas sampling information in Table 4. Record 1) time/date and initials, 2) chamber number, 3) vacuum level of manifold and gas sample bottle before sampling, 4) chamber pressure and temperature before taking sample, and 5) chamber pressure after sampling **WITH CHAMBER VALVE STILL OPEN** (represents final pressure in chamber, manifold, and gas sample bottle).

NOTE: RECORD EVERY GAS REMOVAL EVENT IN TABLE 4, THIS INCLUDES GAS REMOVED IN STEP 17.0 AND 19.0. IT IS IMPORTANT TO KEEP TRACK OF THE TOTAL AMOUNT OF GAS REMOVED AFTER THE TEST STARTS TO DETERMINE TOTAL GAS GENERATED DURING THE TEST.

- 17.2 Figure 5 shows the equipment for taking a gas sample.
- 17.3 Keep the **CHAMBER VALVE CLOSED**.
- 17.4 Connect the manifold to the chamber valve.
- 17.5 Connect the vacuum pump to the manifold.

17.6 Connect one 25 cc gas sample cylinder with attached valve to the manifold. ~~OPEN ONLY THE GAS CYLINDER VALVE AND EVACUATE MANIFOLD~~
MANIFOLD LVF 8/19/93

GAS CYLINDER
17.7 OPEN THE ~~MANIFOLD~~ VALVE to the pump, and evacuate the manifold and gas sample cylinder to less than 50 microns, as indicated by the TC vacuum gauge.

17.8 Record vacuum level. THEN CLOSE MANIFOLD VALVE.

CLOSE MANIFOLD VALVE
TEMPERATURE AND LVF 8/19/93
17.9 Record chamber pressure. THEN OPEN CHAMBER VALVE FULL OPEN.

3 TO 5 LVF 8/19/93
17.10 Wait *3 TO 5* minutes. Note any continuously varying pressures during this time. Record chamber pressure.

17.11 CLOSE CHAMBER VALVE. CLOSE GAS SAMPLE VALVE.

MANIFOLD. GAS SAMPLE VALVE FROM
17.12 Remove ~~gas sample cylinder~~. Remove ~~manifold~~. *MANIFOLD. LVF 8/19/93*

17.13 Take gas sample to wherever Grout Technology designates for gas analysis. Grout Technology shall specify the gases that are to be analyzed.

18.0 ULTRASONIC MEASUREMENT INSTRUCTIONS

Step 18.0 describes ultrasonic measurement for liquid level in the bottom well for each Grout Test Chamber. NDE shall operate the ultrasonic equipment and interpret the CRT screen graphic data. The chambers are sitting on the operating deck along side the gamma irradiation pool for this ultrasonic measurement Step 18.0.

18.1 Record all ultrasonic measurement information in Table 5. Record 1) time/date and initials, 2) chamber number, 3) any interpretation of data given by NDE.

18.2 NDE will place the ultrasonic probe on the bottom of the chamber well and make a determination for the presences and level of liquid.

19.0 PRESSURE EXCEEDS 25 PSIA - GAS REMOVAL FROM GROUT TEST CHAMBER

P in #2 on 10/24/93. Decided to let it ride & pull systems before 2 Atm is exceeded, C.A.H. 11/12/93

Step 19.0 describes the removal of gas from a Grout Test Chamber in the event that the pressure threatens to exceed 25 psia (approximately 10 psig). This Step 19.0 can also be used to reduce the pressure in a chamber at any time C.A. Hinman or L.V. Feigenbutz requests that pressure be reduced.

NOTE: THIS STEP 19.0 SHALL BE PERFORMED UNDER THE DIRECTION OF C.A. HINMAN, OR L.V. FEIGENBUTZ, OR THEIR DESIGNEE.

- 19.1 Whenever the pressure in a Grout Test Chamber exceeds 25 psia, perform the following steps:
- 19.2 Record the removal of 528 cc of gas in Table 4 (this volume includes an estimated volume for the manifold). Record 1) time/date and initials, 2) chamber number, 3) chamber temperature, 4) vacuum level of cylinder before removing the gas, 5) chamber pressure before removing gas, and 6) chamber pressure after removing gas **WITH CHAMBER VALVE STILL OPEN** (represents pressure in chamber and gas cylinder).
- 19.3 Figure 6 shows the equipment for removing gas to reduce the chamber pressure.
- 19.4 Remove the Grout Test Chamber that has the high pressure from the gamma irradiation tube using Step 16.0.
- 19.5 Take one gas sample for gas analysis using Step 17.0.
- 19.6 As shown in Figure 6, connect the calibrated 523.4 cc gas cylinder and small tee manifold to the Grout Test Chamber valve. Connect the vacuum pump and TC vacuum gauge to the manifold valve.
- 19.7 With the **CHAMBER VALVE CLOSED**, open the gas cylinder valve and manifold valve to the pump. Evacuate the gas cylinder and manifold to less than 50 microns.
- 19.8 **CLOSE THE MANIFOLD VALVE.** NOTE - monitor the chamber pressure and **CLOSE THE CHAMBER VALVE** when the chamber pressure is reduced to approximately 15 to 16 psia. **SLOWLY OPEN THE CHAMBER VALVE.**

19.9 CLOSE THE CHAMBER VALVE AND GAS CYLINDER VALVE. Remove the manifold and gas cylinder from the chamber.

19.10 Return the Grout Test Chamber to it's gamma irradiation tube using Step 13.0.

19.11 Dispose of the gas in the gas cylinder per instruction from Grout Technology.

20.0 POST-IRRADIATION CHARACTERIZATION AT 3730 BUILDING

Step 20.0 describes the removal of the six Grout Test Chambers from the irradiation tubes for post-irradiation characterization of the synthetic grout specimens. The characterization will consist of measuring the gas pressures in each of the six Grout Test Chambers for a period of time, to determine if gas continues to be released after removal from the irradiation source. The post-irradiation characterization will be performed on the 3730 Building floor next to the pool.

20.1 HOLD POINT FOR STEP 20.3

TESTING IN THE FOLLOWING GAMMA IRRADIATION TUBES IS COMPLETE AND THE PAIR OF GROUT TEST CHAMBERS IN THIS TUBE SHALL BE REMOVED. APPROVAL SHALL BE SIGNED BY C.A. HINMAN OR HIS DESIGNEE.

TEST IN TUBE 16 COMPLETED: _____ DATE: _____

TEST IN TUBE 17 COMPLETED: _____ DATE: _____

TEST IN TUBE 18 COMPLETED: _____ DATE: _____

CAUTION

20.2 The six Grout Test Chambers are to remain UPRIGHT at all times until disposition Step 21.0 is performed.

20.3 Remove the pair of chambers from the gamma irradiation tube using Step 16.0.

20.4 Take a gas sample from each removed chamber using Step 17.0.

20.5 NDE take a final ultrasonic measurement on each chamber using Step 18.0.

CAUTION

- 20.6 Maintain the integrity of each Grout Test Chamber so that temperature and pressure can be measured at the 3730 Building. DO NOT KINK THE 1/16 INCH TUBING TO THE PRESSURE TRANSDUCER.
- 20.7 Carefully remove all the support tubes from each chamber and support each chamber on the operating deck next to the DAS.
- 20.8 Record on the DAS floppy disk and printer the temperatures and pressures at time intervals and for a length of time specified by C.A. Hinman or his designee.
- 20.9 At the end of this post-irradiation characterization, take one final gas sample from each of the six Grout Test Chambers per Step 17.0.
- 20.10 HOLD POINT FOR STEP 21.0

THE POST-IRRADIATION CHARACTERIZATION IS COMPLETED. THE SIX GROUT TEST CHAMBERS SHALL BE DISPOSITIONED STARTING WITH STEP 21.0. APPROVAL SHALL BE SIGNED BY C.A. HINMAN OR HIS DESIGNEE.

C.A. Hinman

Date

21.0 DISPOSITION OF GROUT SPECIMENS

CAUTION

The six Grout Test Chambers are to remain UPRIGHT at all times until disposition Step 21.10 is performed.

- 21.1 Remove the thermocouple from the top of the chamber and cap the Swagelok fitting.
- 21.2 Remove the pressure transducer tube from the top of the chamber and cap the Swagelok fitting.
- 21.3 Return the six Grout Test Chambers and all equipment to the 306E Building.

21.4 PREPARATION TO BE PERFORMED BEFORE STARTING STEP 21.7

The maximum amount of liquid to accumulate is up to the bottom of the Grout Casting Container in the Grout Test Chamber. This is approximately 200 cc. Obtain 12 or more glass bottles with caps, that have a volume capacity of at least 250 cc. Clean the insides of the bottles and caps with reagent grade acetone and then reagent grade ethanol. Dry each bottle and cap at approximately 100°F under vacuum.

21.5 Label the 12 bottles with the words:

COBALT-60 GROUT IRRADIATION TEST
BOTTLE NO. _____
LIQUID FROM GROUT CHAMBER No. ____

Number the bottles consecutively from 1 through 12. Weigh the 12 empty glass bottles with their caps. Record the bottle numbers and empty weights in the Laboratory Notebook No. WHC-N-763.

21.6 Obtain 12 rubber stoppers, 1/8 inch nominal diameter, for temporarily sealing the Grout Casting Containers.

PERFORM THE FOLLOWING STEPS FOR EACH CHAMBER. ALLOW ENOUGH TIME TO PERFORM STEP 21.7 THRU 21.14 FOR EACH CHAMBER IN ONE WORKING DAY.

21.7 With the chamber maintained in UPRIGHT POSITION AND WITH MINIMUM SHOCK AND VIBRATION, cut off the top cover of the chamber.

CAUTION

DO NOT ALLOW MACHINE CUTTINGS OR CUTTING FLUIDS TO GO INTO THE CHAMBER.

21.8 Remove the top cover and carefully lift the Grout Casting Container out of the chamber BUT KEEP IT POSITIONED OVER THE CHAMBER. While holding the container over the chamber, tilt the container back-and-forth to drain all liquid out of the container into the chamber well.

21.9 Turn the Grout Casting Container over and temporarily plug the 1/8 inch diameter hole with a rubber stopper and set it aside. Cover each chamber to prevent loss of liquid by evaporation.

21.10 HOLD: C.A. HINMAN OR W.J. POWELL IS TO BE PRESENT FOR THIS STEP 21.10 TO ADVISE ON LIQUID REMOVAL AND DISPOSITION.

Carefully drain any liquid in the chamber into one or more of the **WEIGHED** glass bottles, use a syringe and needle if necessary. If there are crystals in the well, remove the crystals with a spatula and put them in the bottle, or remove them as directed by C.A. Hinman or W.J. Powell. Tightly cap the bottles and write the chamber number on the bottles.

21.11 Weigh each liquid-filled bottle and record in the Laboratory Notebook the bottle number, chamber number, and the weight of the bottle.

21.12 Weigh each of the six Grout Casting Containers without the rubber stopper. Record the weights in the Laboratory Notebook. Re-stopper the containers.

21.13 Seal the top lid of each of the six Grout Casting Containers as shown in Figure 7 and as follows:

1. Seal the annulus formed by the 1 inch diameter disk, which is tack welded to the top cover, using the GTAW process. **DO NOT USE A VACUUM TO DO THIS WELD.**
2. GTA weld the 1/4 inch diameter Connecting Tube, item 1 in Figure 7, to the 1/8 inch hole in the top cover.
3. Place a shut-off valve with 1/4 inch Swagelok end fittings on the Connecting Tube, tighten the swagelok fitting and close the valve. The valve shall be rated for 200°F operation.

21.14 Put each sealed Grout Casting Container in a plastic bag and seal the bag.

21.15 Deliver the six Grout Casting Containers and bottles as directed by Grout Technology.