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DURABILITY OF CONTAINERS FOR STORING SOLIDIFIED RADIOACTIVE WASTES*

by

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Abstract

Most concepts for the disposal of highly radioactive waste involve converting the waste to a solid form like concrete or glass and storing this solid form in metal containers. Two major factors in the final selection of materials for these containers are the compatibility between waste form and container material and the durability of the material at temperatures and stresses expected during service and possible accidents. Currently, AISI 1020 carbon steel appears to be a better material than other alloys such as *Cor-Ten A*,** Type 304L stainless steel, or *Inconel* 600*** considered. This choice is based on the results of 10,000 hours of heating tests that showed container compatibility with both concrete and glass waste forms. The selection is also based on 1) analyses of the strengths and 2) oxidation resistances of the alloys under the conditions expected during 100 year storage in air and in various impact and thermal accidents. The thinner wall thickness required for satisfactory performance of the stronger, more-oxidation-resistant alloys is offset by their higher cost per pound.

Introduction

Most concepts for disposing of highly radioactive wastes involve converting waste to a solid form like concrete or glass and storing this solid in metal containers. The concept for managing wastes from Savannah River Plant processes[1] involves sluicing wastes from current storage tanks[2] and separating

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wastes by centrifugation into an insoluble sludge containing ^{90}Sr and ^{239}Pu and a supernate containing ^{137}Cs.[3] The sludge is washed and dried, and the supernate is passed through ion exchange columns to remove >99% of the ^{137}Cs.[4] For the concrete waste form, sludge and cesium-loaded zeolite resin are combined directly with cement and water, and the resulting concrete is cast into metal storage containers.[5] For the glass waste form, sludge, cesium-loaded *Duolite** resin, and borosilicate glass frit are melted either directly in the metal container or in a separate ceramic melter from which the containers are filled continuously.[6] The remaining liquid is evaporated to a solid salt (principally $NaNO_3$) that contains <0.04% of the total biological hazard,[7] and the salt is placed in metal containers for storage. All containers are to be stored in a near-surface facility and cooled by natural convection with unconditioned air until (up to 100 years) they can be transferred to some Federal repository for permanent disposal.

This paper discusses two major factors in selecting materials for the waste containers: compatibility between the waste forms and container materials, and the durability of the materials at the temperatures and stresses expected during service and possible accidents. Compatibility will be assessed from small-scale tests up to 10,000 hours (1.1 years) completed to date. These tests are part of a continuing program to confirm compatibility by heating small capsules of candidate materials containing waste forms up to 50,000 hours at expected service temperatures and also at slightly higher temperatures [up to 350 C (660 F)]. The candidate materials include representatives of carbon and stainless steels and nickel-based superalloys. The tests at the higher temperatures simulate thermal accidents and accelerate any reactions that may occur during the long service life.

Durability of container materials under expected service and possible accident conditions will be assessed from published values for the mechanical and thermal properties of the materials. This assessment serves as a basis for selecting container materials and designs, and for analyzing safety and costs of the waste storage process. For example, where total container weight is a factor, different container sizes will be considered to indicate reduction of costs by reducing the number of containers required. Possible accidents will be considered without regard to their probability or suitability as design bases to indicate the degree of protection against radioactive releases that can be expected from the containers.

Container durability in permanent storage (Federal repository) will not be analyzed until alternative sites are identified and their environments are characterized. Because these environments may be particularly corrosive (for example, from water and H_2S in salt caverns), additional testing will be required. Other steels, such as those used by the petroleum industry, may be appropriate candidates.

Service and Accident Conditions

Conditions expected during filling and storing of containers and possible accidents were reviewed to evaluate materials and assess the durability of containers. Accidents were considered without regard to their probability

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to determine which material properties or design features might be limiting. Container design is important because the effects of many accidents depend on the dimensions of the container.

Service Environments

Container Filling

Conditions expected as the containers are filled are important only for glass waste, because of the high temperature involved [about 1150 C (2100 F)]. Simplified considerations of heat transfer properties indicate that the container will rapidly (in <0.1 hour) heat to 1150 C (2100 F) whether the container is used as a crucible in which the glass is formed ("in-can melting") or a mold in which glass from a continuous melter is cast. The time at the melt temperature will be about 10 hours for in-can melting (based on the conceptual process) and about 1 hour for continuous melting (based on estimated temperature profile in a glass cylinder cooled by natural convection). A stress will be produced in the container wall from the head of liquid glass in the container: about 7×10^{-2} MPa (10 psi) for a nearly full container during in-can melting and about 7×10^{-3} MPa (1 psi) during filling from a continuous melter [0.3 m (1 ft) liquid height].

Pertinent properties of container materials under these conditions are compatibility with molten glass, oxidation resistance, and rupture strength. If an inert-gas shroud or other protection is not provided, oxidation will thin the container wall; spalled oxide can constitute a maintenance problem. Because rupture of a container is unacceptable, the rupture stresses at 1150 C (2100 F) for both 1 and 10 hours respectively, will be considered the critical mechanical properties.

Some permanent deformation of the container wall will occur because the thermal expansion of steel is greater than that of glass. Most of the stress resulting from these differences in thermal expansion will be relaxed during cooling from melting temperatures.

Container Storage

The expected environment for the containers during 100 year storage is unconditioned air. The container surface will be at approximately 100 C (212 F), depending on the final design of the container and the storage building. Principal factors affecting container life are compatibility with the waste form, corrosion of the external surface, and stress in the walls from pressures inside the containers. Because the containers are to be cooled by natural convection with unconditioned air, resistance to atmospheric corrosion is a pertinent material property. Internal pressure will cause creep. To conservatively avoid extensive deformation of the container, the stress required to produce 1% creep during 100 years (8.76×10^6 hours) will be a pertinent mechanical property.

Three sources of pressure inside a concrete-filled container are the following: air inside the freeboard space will expand as it heats to the storage temperature, steam will be liberated from the concrete, and O_2 and H_2 will be generated by radiolysis. The contribution of air pressure to total pressure will depend on conditions at the time the container is sealed; however, at the storage temperature [100 C (212 F)], pressure will not be much greater than one atmosphere [0.1 MPa (15 psi)].

The steam pressure in a container will correspond to the equilibrium value given in standard steam tables, as shown by small-scale tests.[5] In these tests, a small concrete cylinder was sealed in a container equipped to monitor continuously the temperature and pressure in the void space above the concrete. The unit was heated in various temperature stages up to 240 C (465 F). For each temperature stage, the pressure rose much more slowly than the temperature, but the pressure ultimately reached the equilibrium predicted by steam tables. For example, about 2 hours was required for heating the unit from 200 C (392 F) to 240 C (465 F) and for the temperature to reach equilibrium. About 24 hours was required for the pressure to reach equilibrium. This time delay probably represents the time required for the steam to diffuse out of the concrete and to saturate the void space.

During storage, radiolysis will generate O_2 from nitrate in the waste and H_2 from water, or any organics present.[8] With data for the conceptual process, calculated O_2 and H_2 pressures at 100 C (212 F) would be 2.2 MPa (315 psi) and 0.2 MPa (25 psi), respectively. No adverse effects on mechanical properties of the container material are expected from this small amount of hydrogen. The indicated oxygen pressure may never be attained because oxidation of the inner surface of the container will consume a large portion of this oxygen. However, no credit for this effect will be taken in the analysis.

The only identifiable source of pressure in a glass-filled container during storage is the expansion of the air inside. As indicated above, a pressure of 0.1 MPa (15 psi) at 100 C (212 F) will be assumed.

Accident Environments

Possible accidents can be divided into two categories: thermal accidents, such as fire or loss of cooling in the storage building, and impact accidents, such as dropping a container from a crane or truck or being hit by a high velocity missile. Thermal events cause increased reaction between waste form and container, oxidation of container surfaces, and increased internal pressure. Impact accidents cause plastic deformation and possibly penetration of the container wall.

Thermal Accidents

In the unlikely event cooling is lost in the storage building, the container temperature is expected to be about 325 C (615 F).[9] Because of the expected design of the building, 30 days (720 hours) is a conservative estimate of the delay before normal cooling is restored. Corresponding internal pressures would be 16 MPa (2400 psi) for a concrete-filled container and 0.2 MPa (25 psi) for a glass-filled container. The stress required to produce 1% creep during 720 hours at 325 C (615 F) is used as the critical mechanical property, assuming that some deformation is tolerable; rupture is unacceptable because of the large number of containers involved.

The estimated condition resulting from a fire is a container wall temperature of 790 C (1450 F) for 0.5 hour with internal pressures of 27.5 MPa (4000 psi) for a concrete-filled container or 0.3 MPa (45 psi) for a glass-filled container. These estimates are based on the same conditions [flame temperature of 790 C (1450 F) which lasts for 0.5 hour] which are used to analyze effects of fire on shipping casks for radioactive materials.[9] From a simplified analysis of nonsteady-state heat transfer in a 0.6 m (2 feet) diameter container with 1.25 cm (0.5 inch) thick wall, the wall and the air-filled freeboard are expected to reach the fire temperature in about 0.5 hour, but only the outer surface of the waste form (>R/2) would experience any temperature rise. The pressure contributions from air [0.3 MPa (45 psi)] and radiolytic gases [6.6 MPa (965 psi)] were based on their expansion as they heated to the fire temperature. The pressure contribution from steam in a concrete-filled container [20.6 MPa (2995 psi)] corresponds to the equilibrium temperature calculated on an energy balance basis 370 C (700 F).

The actual pressures attained may be less than the above estimates for two reasons: 1) the duration of the fire may be short enough that the equilibrium pressure would not be attained because of the time delay that was observed in the heating tests described above, and 2) the increased internal volume of the container resulting from thermal expansion would reduce the pressure. However, no credit for these effects will be taken in this analysis.

Impact Accidents

A number of empirical equations to predict penetration on impact have been developed for different applications; design-basis accidents to shipping casks for radioactive materials,[9] response of nuclear reactor tanks to tornado-generated missiles,[10,11] and high-velocity ballistic impact technology.[12,13] These equations are similar because they relate penetration to the kinetic energy of impact. The relation developed for cask design [a 1 m (40 inches) drop on a 15 cm (6 inches) diameter unyielding peg] is used in the present analysis because the impact energy for typical waste containers can be compared to that for typical high-velocity missiles [3 m (10 feet) long section of 7.6 cm (3 inches) diameter, Schedule 40 steel pipe traveling at 160 km/hr (100 mph)].

Candidate Container Materials

Five alloys are considered: AISI 1020 carbon steel, *Cor-Ten A*, Type 304L stainless steel, *Inconel* 600, and *Inconel* 625. *Cor-Ten A* is a low-alloy steel containing 1% chromium, 0.5% nickel, and 0.35% copper that is noted for its resistance to atmospheric corrosion. Data on the creep strength of *Cor-Ten A* or alloys of similar composition are limited but suggest that strength at elevated temperatures is similar to that of AISI 1020. The two *Inconel* alloys are nickel-based superalloys known for their strengths and resistances to oxidation at high temperatures. These five commercially available alloys represent materials expected to be suitable for waste containers and, except for *Inconel* 625, are included in the matrix of compatibility tests. Other steels and superalloys [such as 18 Ni(300), a maraging steel, and *René* 41*] offer no particular advantage because their very high strengths are not maintained at the elevated temperatures expected in accidents. Other alloys, such as those based on aluminum, can be eliminated from consideration for one or more reasons: cost, expected incompatibility with concrete, or low melting point.

Compatibility

Compatibility between the candidate container materials and waste forms is being measured by heating small capsules of the materials containing either real or synthetic (nonradioactive) wastes up to 50,000 hours (5.7 years) at expected service temperatures and at slightly higher temperatures [up to 350 C (660 F)]. In most cases, five capsules of each combination of material and waste form were fabricated and placed in furnaces for heating. After desired intervals (usually 1000, 5000, and 10,000 hours), one capsule of each type was removed from the furnace, sectioned, and examined metallographically for reactions at the waste-capsule interface.

To study the concrete waste form, several sets of samples representing combinations of real and synthetic sludge, Type I portland and *Lumnite*** (high alumina) cements, and sealed and unsealed capsules were made. To test the corrossiveness of the "caustic" steam generated from heated concrete, sealed capsules were fabricated from 1 cm (3/8 inch) (NPS) high-pressure, Type 304L stainless steel pipe fittings. A pipe plug was welded into one end of a coupling and the cavity was partially filled with concrete made from synthetic sludge and each of the two types of cement. A sample of *Cor-Ten A* or 304L stainless steel made by bending 0.25 cm (0.1 inch) diameter rod into the shape of a paper clip was placed so that one U-bend was in the concrete and the other U-bend was in the vapor space above. After curing the concrete for one day, a plug was welded in the other end of the coupling. These capsules were heated at 100 and 300 C (212 and 570F), creating internal steam pressures of 0.1 MPa and 14 MPa (15 psi and 2000 psi), respectively.

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* Trademark of General Electric Company.

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Unsealed capsules were fabricated from each of the candidate materials and were filled with each combination of real and synthetic sludge in each type of cement. After curing one day, a disc-shaped lid was placed on top of the capsules. These capsules were 1.25 cm (0.5 inch) diameter by 1.25 cm (0.5 inch) high with a cavity 0.5 cm (0.188 inch) diameter by 0.5 cm (0.188 inch) deep for the concrete. These capsules were heated at 100 and 350 C (212 and 660 F).

To study the glass waste form, synthetic sludge, zeolite, and borosilicate glass frit were melted together in a ceramic crucible; the glass was cast into capsules similar to the unsealed capsules used for the concrete tests. Two sets of capsules, one clean and one preoxidized by heating it in air for 1 hour at 1000 C (1830 F), were examined metallographically without any long-term heating. These two sets of capsules were examined to separate the effects of the molten glass.

To study in-can melting of glass, about 5 cm (2.0 inches) diameter by 61 cm (24 inches) long crucibles were fabricated from 304L stainless steel and *Cor-Ten*.[14] Synthetic sludge and glass frit sufficient to fill the crucible with molten glass to a depth of 15 cm (6 inches) were blended together and divided into three portions. The first portion was heated in the crucible to 1150 C (2100 F); the other two portions were each added and melted. After holding the melt for 3 hours, the crucible was transferred to another furnace at 500 C (930 F) and allowed to cool slowly. The crucible and glass contents were sectioned at several elevations, and the crucible wall was examined for any thinning.

Unsealed capsules were also tested for reactions with synthetic waste components: dehydrated supernate (dried with a wiped-film evaporator), sludge, and cesium-loaded zeolite. The sludge and cesium-loaded zeolite were tested because these components in the concrete waste form would inevitably be in direct contact with the container at several locations.

Mechanical and Physical Properties

The costs and pertinent physical and mechanical properties of these five alloys are summarized in Table 1.[15-19] Values for creep and rupture strengths were extrapolated from the literature data with the Larsen-Miller parameter. This parameter relates the time and temperature of stress application to the allowable deformation or rupture.[20] Typical examples of Larsen-Miller curves are shown in Figure 1.

Material Behavior in Service and Accident Environments

Compatibility

Completed tests (up to 10,000 hours) have demonstrated satisfactory compatibility between each of the waste forms and the candidate container materials. Changes in wall thicknesses of the capsules were negligible; effects of the long-term heating were detected only by microscopic examination.

Unsealed capsules containing concrete waste made from synthetic and real sludges were examined after the synthetic sludges were heated for 10,000 hours and the real sludges were heated for 5000 hours. Heating produced continuous reaction layers up to 0.25 μm (0.0001 inch) thick on all materials. Infrequent patches of intergranular penetration up to 7.5 μm (0.003 inch) deep occurred only on AISI 1020 and *Cor-Ten A* (Figure 2). The thinness of the continuous reaction layers precluded any detailed analysis of growth kinetics. However, the layer was thinner at 100 C (212 F) than at 350 C (660 F), and the more oxidation-resistant alloys (*Inconel* and 304L stainless steel) had the thinnest reaction layers. Similarly, the intergranular penetration of AISI 1020 and *Cor-Ten A* was observed at 350 C (660 F) after 5000 hours and at both 100 and 350 C (212 F and 660 F) after 10,000 hours. No compatibility differences between synthetic and real sludges have been observed.

Heating the sealed capsules for 1000 hours produced a continuous reaction layer adjacent to the concrete that was thicker than in unsealed capsules, but no intergranular attack was detected. At 100 C (212 F), the layer was <0.25 μm (0.0001 inch) thick on both *Cor-Ten A* and 304L stainless steel; but at 300 C (570 F), the layer was 2.0 μm (0.0008 inch) thick on *Cor-Ten A* and 0.75 μm (0.0003 inch) thick on 304L stainless steel. The steam produced a reaction layer 0.25 μm (0.0001 inch) thick and a surface deposit of plate-shaped crystals (Figure 3). No cracking was seen in the stressed (U-bend) portions of the samples.

Capsules which contained vitrified synthetic waste were heated at 100 and 350 C (212 F and 660 F) for up to 10,000 hours. When these capsules were examined, no reaction with the glass had occurred during the long-term heating. A thin <1.3 μm (0.0005 inch) oxide layer was present adjacent to the glass. However, the thickness of the layer on capsules heated at 100 C (212 F) was the same as on those heated at 350 C (660 F). Tests with preoxidized capsules showed that the oxide did not dissolve or spall during the filling operation.

Tests of the in-can melting concept indicated that 304L stainless steel might be a satisfactory material, but *Cor-Ten* would not be satisfactory, as shown by the reductions in wall thickness given in Table 2. The reaction with molten glass resulted primarily because oxide that formed on the inside of the can during heating and melting of the waste and glass powders subsequently dissolved in the molten glass. As the glass cooled, a band of dendritic-shaped particles, identified by x-ray diffraction as γ-Fe_2O_3, precipitated in the glass near the can surface (Figure 4). No sensitization of the 304L stainless steel was observed in these tests. However, sensitization might occur in a full-sized container depending on the exact temperature history.

Capsules containing dried synthetic supernate, sludge, or cesium-loaded zeolite have only shown light discoloration (304L stainless steel, yellow; AISI 1020 and *Cor-Ten A*, brown) after they were heated up to 10,000 hours. The contents of the capsules were easily dumped or washed out of the capsules. No dimensional changes occurred in the capsules, and metallographic examination showed no intergranular attack or pitting. Microprobe analysis of the capsule that contained cesium-loaded zeolite did not show any concentration of cesium (<0.3 wt %) at the inner capsule surface.

These data were used to estimate the wall thickness affected by compatibility reactions at the expected service and accident conditions. For concrete, the observed reaction with AISI 1020 steel and *Cor-Ten* extrapolates to 87.5 μm (0.035 inch) after 100 years, assuming a parabolic rate and 12.5 μm (0.005 inch) for 304L stainless steel and *Inconels*. For glass, the thickness affected was assumed to be the same as that consumed by oxidation because the only reactions observed were associated with the oxide. No allowance was used for thermal accidents because the affected thickness was negligible after 10,000 hours at 350 C (660 F), and the expected duration of accidents is much shorter.

Atmospheric Corrosion

Under the expected storage conditions [100 years at 100 C (212 F)], the exterior container surface will oxidize at a rate that can be estimated from data on the atmospheric corrosion of the representative alloys or ones of similar composition.[21-24] These data are reported as weight lost from which a uniform penetration was calculated for exposures to rural, industrial, and marine atmospheres for up to 15 years. To provide conservative estimates of corrosion resistance, data for the more-aggressive marine atmosphere were used. The penetration increased parabolically with time (Figure 5) as expected for an oxidation reaction and as observed for high temperature oxidation in air.[25] Because this relationship is observed for these extreme conditions, the data can be extrapolated toward 100 year exposures with confidence.

Uniform penetration in 100 years of 0.13 to 0.25 cm (0.05 to 0.1 inch) would be expected for AISI 1020, 0.025 to 0.05 cm (0.01 to 0.02 inch) for *Cor-Ten A*, and <0.0025 cm (0.001 inch) for 304L stainless steel and the *Inconels*. Pits two to three times deeper than the uniform attack would also be expected;[22] such pitting should have little effect on mechanical properties. Although no quantitative data are available for 304L stainless steel, the expected penetration is equal to or less than that observed for *Inconel* 600. Inspections of the 300 series stainless steels used on the exteriors of the Empire State and Chrysler Buildings in New York City showed only pitting attack of a few microns deep after 20 to 30 years.[26]

High Temperature Oxidation

At the high temperatures associated with service and accident conditions, the exterior surface of the container will oxidize. The extent of oxidation can be evaluated from literature data shown in Figure 6. These data show the uniform penetration measured after 1000 hour exposures at various temperatures. The values for carbon steel (representative of AISI 1020 and *Cor-Ten A*) and 304L stainless steel were calculated from measured weight losses;[27,28] actual depths of penetration may be slightly larger depending on the contributions of pitting and grain boundary oxidation. The data for *Inconel* 600 are from metallographic measurements that include grain boundary effects.[25] Oxidation for times other than 1000 hours can be calculated from the observed parabolic rate of oxidation.

Significant [>0.025 cm (0.01 inch)] oxidation of any of the alloys would be expected only during the filling of the containers with glass. The oxide layer on carbon or stainless steel containers would be 0.075 to 0.114 cm (0.030 to 0.045 inch) thick for in-can melting and 0.025 to 0.064 cm (0.010 to 0.025 inch) for continuous melting. On the *Inconels*, the oxide layer would be about 0.013 cm (0.005 inch). Most of this layer would spall off as the containers cooled.* Alternatively, a blanket of flowing gas may reduce this oxidation.

Mechanical Stresses

To assess the response of container materials to various mechanical stresses of service and accident environments, equations that relate the dimensions of the containers to conventional properties, such as density and tensile, creep, and rupture strengths were developed. These derivations result from simultaneous solution of the equations for the hoop (circumferential) stress in the container wall, thickness of container wall required to withstand a 1 m (40 inches) drop onto a peg, and volumes of steel and waste in a container.

A condition for survival of containers in different environments can be expressed by equating hoop stress to the allowable creep or rupture stress, σ_c. The desired wall thickness t is

$$t = \frac{pR}{\sigma_c} \qquad (1)$$

where p is the internal pressure, and R is the inside radius of the container. The value of t is substituted in the expression for the ratio of the volume of container V_s to the volume of waste V_w. This substitution gives

$$\frac{V_s}{V_w} = \frac{\pi h(2Rt+t^2)}{\pi hR^2} = \frac{2Rt+t^2}{R^2}$$

$$\frac{V_s}{V_w} = \frac{p}{\sigma_c}\left(2+\frac{p}{\sigma_c}\right) \qquad (2)$$

The empirical drop-test equation is

$$t_{min} = \left(\frac{W}{\sigma_\mu}\right)^{0.71}$$

* A 0.114 cm (0.045 inch) thick layer on a 0.61 m (2 feet) diameter by 3 m (10 feet) high container corresponds to 6800 cm^3 (0.24 ft^3) or 35 kg (77 pounds).

where t_{min} is the minimum wall thickness required to prevent penetration, W is the total weight, and σ_μ is the ultimate tensile strength of the container material.[10] W is expressed in terms of the volumes and densities of container and waste form. Equation 2 gives

$$\left[t_{(min)}\right]^{1.41} = V_w \left[\frac{\rho_w \dfrac{V_s}{V_w} + \rho_w}{\sigma_\mu}\right]$$

$$\left[t_{(min)}\right]^{1.41} = V_w \frac{\rho_s p^2 + 2\rho_s p\sigma_c + \rho_w \sigma_c^2}{\sigma_\mu \sigma_c^2} \qquad (3)$$

where ρ_s is the density of the container, and ρ_w is the density of the waste form.

For design purposes, safety factors on both the allowable stress and the wall thickness are desirable. The hoop stress should be some fraction a of the creep or rupture strength. Equation 1 gives

$$t = \frac{pR}{a\sigma_c} \qquad (4)$$

The desired wall thickness should be some multiple b of t_{min}, and Equation 3 gives

$$t^{1.41} = b^{1.41} A\, V_w \qquad (5)$$

where

$$A = \frac{\rho_s p^2 + 2\rho_s p\sigma_c + \rho_w \sigma_c^2}{\sigma_\mu \sigma_c^2} \qquad (6)$$

Equations 4 and 5 and the equation for V_w are plotted in Figure 7.

These relationships were illustrated by evaluating the dimensions of an AISI 1020 steel container for storage of concrete waste. Appropriate values for the material parameters A and p/σ_c were calculated for the storage conditions [100 years at 100 C (212 F)] and corrected for an assumed safety factor of 50% (a = 0.5 and b = 1.5). With these values, Figure 7 gives corresponding values of t, R, and h that satisfy the relationships. For example, the resulting container has a wall thickness of 0.5 cm (0.2 inch), a radius of 42 cm (16.7 inches), a height of 1.6 m (5.2 feet), and a capacity of 0.85 m^3 (30 ft^3).

The minimum wall thicknesses required for adequate strength in each service or accident environment were calculated for a container with 0.61 m (2 feet) diameter by 3 m (10 feet) high (Table 3). For each environment, different strengths (σ_c) and safety factors (a) were used for the effect of internal pressure to reflect "reasonable" limits for deformation of the container wall. For example, for storage, the stress for 1% creep and a safety factor of 0.5 were required to indicate that little deformation could be tolerated. Because a large number of containers would be affected by a loss of cooling accident, the stress for 1% creep was used, but a safety factor a = 1 was assumed because moderate deformation of the containers could be accepted as long as they did not rupture. In glass melting, some deformation could be tolerated, but not rupture; therefore, the rupture strength and a safety factor of 0.5 were used. A constant safety factor of b = 1.5 was used for impact accidents.

These calculated wall thicknesses indicate that impact accidents are generally the most important considerations, except for the high pressures developed in thermal accidents to concrete-filled containers, or for the high temperatures required for in-can melting of glass.

Evaluation of Container Materials and Waste Forms

The container materials and waste forms were compared by calculating a "figure of merit" (M) for each material in the different service and accident environments (Table 4). Each value for M is the reciprocal of the cost of the metal in a 0.61 m (2 feet) diameter and 3 m (10 feet) high container that has the minimum wall thickness required to withstand the expected conditions. This wall thickness (t, in Table 4) is the sum of the thicknesses required for strength (Table 3), atmospheric corrosion (Figure 5), high temperature oxidation (Figure 6), and compatibility. Current prices for 1.3 cm (0.5 inch) thick plate were used to reflect the cost of container materials.

For any combination of waste form and service or accident conditions, a carbon steel container with sufficiently thick walls to withstand expected stresses and reactions is better than any of the other materials by a factor of two, or more. Comparison of the values for wall thickness (t) and merit (M) shows that the higher strengths and corrosion resistances of stainless steel and the *Inconels* are offset by their higher costs. 304L stainless steel is equivalent to carbon steel only in the cases of a concrete-filled container in a fire and in-can melting of glass.

The merit values also provide one of many bases for evaluating the waste forms. This selection largely depends on which service and accident conditions are ultimately selected as the bases for final design. For example, if loading and storage of containers are the only criteria, concrete has slightly higher merit than continuously melted glass and considerably higher merit than in-can melted glass. However, essentially the same container that is required for storing continuously melted glass will also withstand accidents, such as loss of cooling and fire. This anticipated durability under severe conditions may outweigh the greater investment in containers (ratio of M-values = 6.6/4.9 = 1.35).

These considerations also indicate that double containment should not be required solely for durability during 100 year storage. However, a secondary container may be required for some other considerations, such as providing a contamination-free outer surface or durability in the environment of final disposal (Federal repository).

Many other factors can affect choice of waste form and container dimensions. As an example, container capacities as large as possible would be desirable to reduce the number and total cost of containers. A minimum wall thickness [for example, 1.3 cm (0.5 inch)] may be required for rigidity in handling empty containers and welding the final closure. Radius may be limited to some maximum to prevent excessive centerline temperatures.* With the above analysis, a container for storage of concrete that would meet these additional criteria would be 2.74 m (9 feet) high with a radius of 0.61 m (2 feet) and a wall thickness of 1.3 cm (0.5 inch), weighing 6630 kg (7.3 tons) and providing a capacity of 3.2 m^3 (113 ft^3). The safety factors are a = 0.3 and b = 1.5. A similar container, but with walls 1.9 cm (0.74 inch) thick and weighing 10,400 kg (11.5 tons), would be satisfactory for continuously melted glass during storage and fire (safety factors a = 0.42 and b = 1.5). The thicker wall is required mainly because the density of glass is greater than that of concrete. Ultimately container dimensions may be limited by the size of the cask used for shipping containers to the permanent storage site.

Summary

When the cost of materials for waste containers is included, 1020 carbon steel appears to be a better candidate than any of the other alloys considered: *Cor-Ten A*, Type 304L stainless steel, *Inconel* 600, and *Inconel* 625. This choice is based on the oxidation resistances, strengths, and compatibilities of the alloys under the conditions expected during 100 year storage, melting of glass by either in-can or continuous-melter processes, and impact and thermal accidents, such as loss of cooling and fires. The thinner wall thickness required for satisfactory performance of the stronger, more-oxidation-resistant alloys such as 304L stainless steel is offset by their higher cost per pound.

A 0.61 cm (2 feet) diameter by 3 m (10 feet) high carbon steel container with a wall thickness about 1.3 cm (0.5 inch) would be expected to survive most service and accident environments with either a concrete or glass waste form. In this analysis, survival is conservatively defined as <1% deformation by creep during storage, about 1% creep in a loss of cooling accident, no penetration in an impact accident, and creep >1% (but no rupture) in a fire. Because of the high internal pressures associated with concrete-filled containers in thermal accidents, and the high temperatures associated with in-can melting of glass, much thicker walls [up to 25.4 cm (10 inch)] would be required to survive these conditions. These alternatives show the strong dependence of the choice of waste form on the accident conditions which are finally selected as the bases for design of the waste management process.

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* Assuming surface temperatures of 100 C (212 F) and maximum centerline temperatures of 250 C (480 F) for concrete and 500 C (930 F) for glass, the maximum radius for transfer of heat from radioactive decay is 84 cm (2.75 feet) for concrete and >91 cm (3.0 feet) for glass.

References

1. Wallace, R. M., Hale, W. H., Bradley, R. F., Hull, H. L., Kelley, J. A., Stone, J. A., Thompson, G. H. "High-Level Radioactive Waste Management," *Advances in Chemistry Series No. 153*, American Chemical Society, Washington, DC (1976).

2. Hill, Jr., A. J. *Removal of Sludge from High Activity Waste Tanks*, USAEC Report DP-1093, E. I. du Pont de Nemours and Company, Savannah River Laboratory, Aiken, SC (1967).

3. Stone, J. A. *Separation of SRP Waste Sludge and Supernate*, ERDA Report DP-1441, E. I. du Pont de Nemours and Company, Savannah River Laboratory, Aiken, SC (to be published).

4. Wiley, J. R. *Decontamination of SRP Waste Supernate*, ERDA Report DP-1436, E. I. du Pont de Nemours and Company, Savannah River Laboratory, Aiken, SC (1976).

5. Stone, J. A. *Evaluation of Concrete as a Matrix for Solidification of Savannah River Plant Waste*, ERDA Report DP-1448, E. I. du Pont de Nemours and Company, Savannah River Laboratory, Aiken, SC (to be published).

6. Kelley, J. A. *Evaluation of Glass as a Matrix for Solidification of Savannah River Plant Waste - Radioactive Studies*, ERDA Report DP-1397, E. I. du Pont de Nemours and Company, Savannah River Laboratory, Aiken, SC (1975).

7. Goodlett, C. B. "Concentration of Aqueous Radioactive Waste with Wipeafilm Evaporators," *Chem. Eng. Progr.*, *72*, 63 (1976).

8. Bibler, N. E. "Radiolytic Gas Production During Long-Term Storage of Nuclear Wastes" to be presented at the *28th Southeastern Regional Meeting of the American Chemical Society* in Gatlinburg, TN, October 27-29, 1976.

9. Shappert , L. B. *Cask Designers Guide: A Guide for the Design, Fabrication and Operation of Shipping Casks for Nuclear Applications*, ORNL-NSIC-68 (February 1970).

10. Russell, C. R. *Reactor Safeguards*, Pergamon Press, New York (1962).

11. Nichols, C. M., Ed. *Progress in Nuclear Energy IV, Technology Engineering and Safety*, Pergamon Press, New York (1961).

12. Byrnside, N. C., Torvik, P. J., Swift, H. F. "Impact Crater Formation at Intermediate Velocities," *Trans. ASME*, *94D*, 395 (1972).

13. Rinehart, J. S., Pearson, J. *Behavior of Metals Under Impulsive Loads*, American Society for Metals, Cleveland, OH (1954).

14. Rankin, W. N., Kelley, J. A. *Correlation of Radionuclide Leachabilities with Microstructures of Glasses Containing Savannah River Plant Waste*, ERDA Report DP-1411, E. I. du Pont de Nemours and Company, Savannah River Laboratory, Aiken, SC (1976).

15. *Alloy Digest*, Engineering Alloys Digest, Inc., Upper Montclair, NJ.

16. *Engineering Properties of Inconel Alloy 625*, Tech. Bull. T-42, The International Nickel Company, Huntington, WV (1966).

17. *Engineering Properties of Inconel Alloy 600*, Tech. Bull. T-7, The International Nickel Company, Huntington, WV (1964).

18. Miller, R. F., Heger, J. J. *Report on the Strength of Wrought Steels at Elevated Temperatures*, STP-100, American Society for Testing Materials, Philadelphia, PA (1957).

19. Simmons, W. F., Cross, H. C. *Report on Elevated-Temperature Properties of Wrought Medium-Carbon Alloy Steels*, STP-199, American Society for Testing Materials, Philadelphia, PA (1957).

20. Conway, J. B. *Stress-Rupture Parameters: Origin, Calculation and Use*, Gordon and Breach Science Publishers, New York (1969).

21. Copson, H. R. "Long-Time Atmospheric Corrosion Tests on Low-Alloy Steels." *Proc. ASTM, 60*, 650 (1960).

22. Mannweiler, G. B. "Corrosion Test Results on Fifteen Ferrous Metals After Seven-Years Atmospheric Exposure," *Metal Corrosion in the Atmosphere*, STP-435, American Society for Testing Materials, Philadelphia, PA, p 211 (1968).

23. Gerhold, W. F. "Report of Subcommittee XVI on Field Tests of Atmospheric Corrosion of Hardware," *Proc. ASTM, 70*, 80 (1970).

24. van Rooven, D., Copson, H. R. "Atmospheric Corrosion Behavior of Some Nickel Alloys," *Metal Corrosion in the Atmosphere*, STP-435, American Society for Testing Materials, p 175 (1968).

25. Angerman, C. L. "Long-Term Oxidation of Superalloys," *Oxid. Metals, 5*, 149 (1972).

26. Mayne, C. R. "Report on Inspection of Corrosion-Resistant Steels in Architectural and Structural Applications," *Proc. ASTM, 61*, 188 (1961).

27. Day, M. J., Smith, G. V. "Iron Alloy Scaling," *Ind. Eng. Chem., 35*, 1098 (1943).

28. Miller, R. F., Benz, W. G., Day, M. J. "Creep Strength, Stability of Microstructure and Oxidation Resistance of Cr-Mo and 18 Cr-8 Ni Steels," *Trans. ASM, 32*, 381 (1944).

TABLE 1 - Properties of Candidate Container Materials

Property	*1020 CS*	*Cor-Ten A*	*304L SS*	*Inconel 600*	*Inconel 625*
Density, lb/in^3	0.284	0.283	0.290	0.304	0.305
Melting point, °C	1515	1510	1455	1425	1285
Tensile strength at 25°C, psi	65,000	70,000	75,000	90,000	120,000
Yield strength at 25°C, psi	38,000	50,000	28,000	36,000	60,000
Stress for 1% creep, psi					
Glass melting (10 hr at 1150°C)	<100	*a*	~100	~250	~500
Storage (8.76×10^5 hr at 100°C)	60,000	-	60,000	75,000	100,000
Loss of cooling (720 hr at 825°C)	40,000	-	55,000	70,000	90,000
Fire (0.5 hr at 790°C)	2,000	-	13,000	5,000	30,000
Rupture strength, psi					
Glass melting - "In-can" (10 hr)	~100	-	1000	1000	~1000
- Continuous (1 hr)	~200	-	2000	2000	~1500
Storage	65,000	-	65,000	90,000	12,000
Loss of cooling	45,000	-	60,000	85,000	100,000
Fire	3,500	-	20,000	15,000	50,000
Cost, $/b	0.18	0.35	0.95	4.00	5.50

a. Creep and rupture properties similar to 1020 CS.

TABLE 2 - "In-Can" Melter Reactions

	Wall Thinning, mils	
	Cor-Ten	*304L SS*
Outer surface		
Oxidation	30	1
Inner surface		
Sludge 13H	53	<1
Sludge 3	96	2

TABLE 3 - Wall Thicknesses Required for Strength

Environment	*Strength Criteria for Response to Internal Pressure*	*Minimum Wall Thickness, in.*[a]			
		1020 CS	*304L SS*	*Inconel 600*	*Inconel 625*
Concrete					
Storage	1% creep, a = 0.5	0.22[c]	0.20[c]	0.17[c]	0.14[c]
Loss of cooling	1% creep, a = 1	0.70[b]	0.51[b]	0.40[b]	0.32[b]
Fire	rupture, a = 1	10[b]	2.5[b]	3.4[b]	0.98[b]
Glass					
Melting - "In-Can"	rupture, a = 0.5	2.5[b]	0.61[b]	0.98[b]	0.24[b]
- Continuous	rupture, a = 0.5	0.30[c]	0.26[c]	0.23[c]	0.19[c]
Storage	1% creep, a = 0.5	0.30[c]	0.26[c]	0.23[c]	0.19[c]
Loss of Cooling	1% creep, a = 1	0.30[c]	0.26[c]	0.23[c]	0.19[c]
Fire	rupture, a = 1	0.31[c]	0.27[c]	0.23[c]	0.19[c]
Fire	1% creep, a = 1	0.32[c]	0.27[c]	0.24[c]	0.20[c]

a. Assumed reference design container, R = 1 ft, h = 10 ft, V_w = 31.4 ft^3; safety factor for impact, b, = 1.5.

b. Thickness is that required to resist deformation from internal pressure, but greater than that required for impact resistance.

c. Thickness is that required for impact resistance, but greater than that required for internal pressure.

TABLE 4 - Relative Merit of Container Materials

Environment	*1020 CS* t^a	*1020 CS* M^b	*Cor-Ten A* t	*Cor-Ten A* M	*304L SS* t	*304L SS* M	*Inconel 600* t	*Inconel 600* M	*Inconel 625* t	*Inconel 625* m
Concrete										
Storage[c]	0.325	6.6	0.270	4.1	0.205	2.0	0.175	0.51	0.145	0.45
Loss of cooling[d]	0.805	2.6	0.750	1.4	0.515	0.76	0.405	0.22	0.325	0.20
Fire[e]	>10	0.15	>10	0.079	2.505	0.15	3.405	0.023	0.985	0.064
Glass										
"In-can melting" and storage[c]	2.650	0.74	2.595	0.38	0.670	0.58	0.990	0.088	0.250	0.26
Continuous melting and storage[c]	0.410	5.2	0.355	3.1	0.280	1.4	0.230	0.39	0.190	0.34
Loss of cooling[d]	0.410	5.2	0.355	3.1	0.280	1.4	0.230	0.39	0.190	0.34
Fire[e]	0.420	5.0	0.365	3.0	0.290	1.3	0.230	0.39	0.190	0.34
Fire[d]	0.430	4.9	0.375	3.0	0.290	1.3	0.240	0.37	0.200	0.33

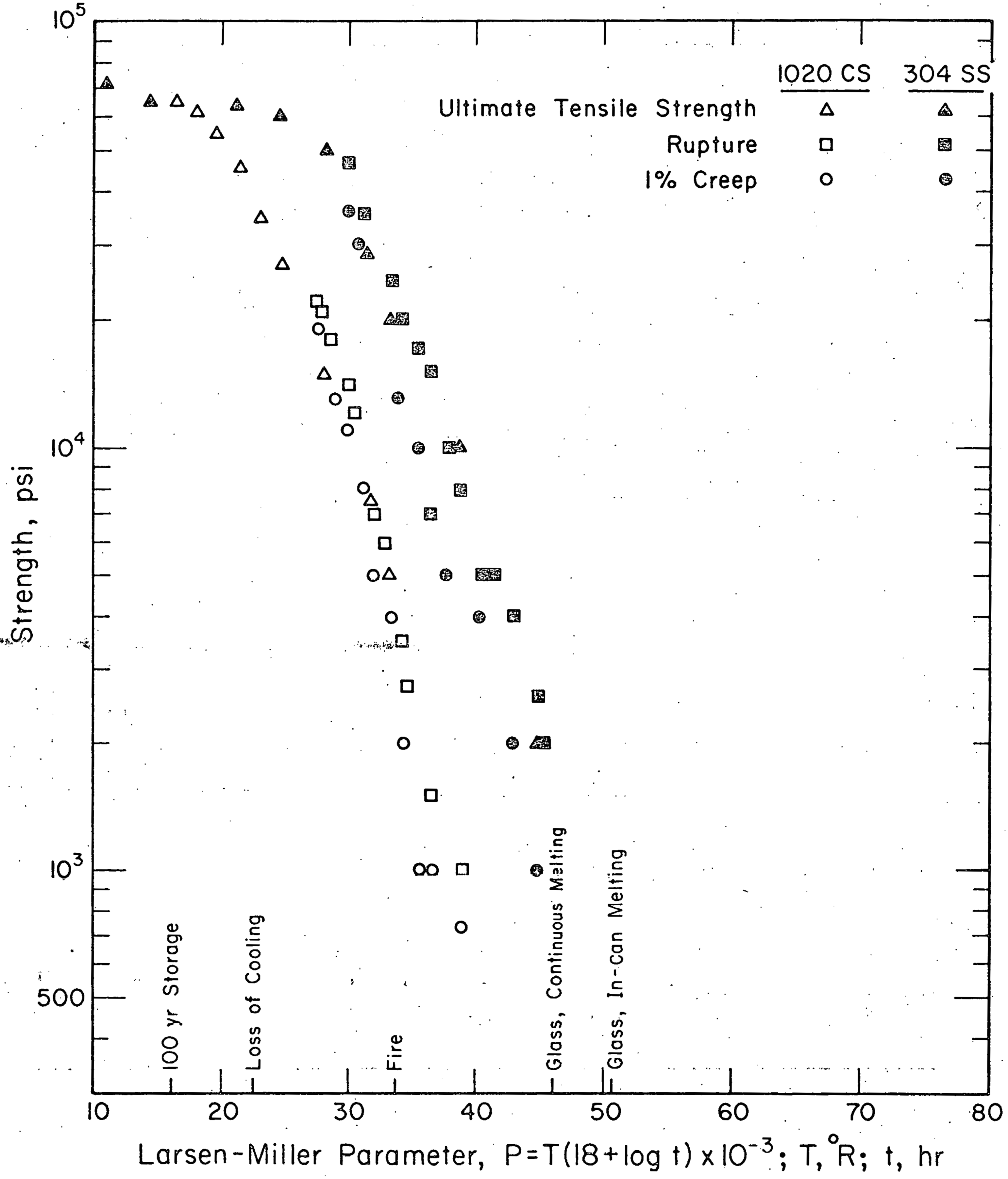
a. t = sum of wall thickness allowances for strength (Table 3) and oxidation (Figures 5 and 6) and compatibility (see text).

b. $M = \frac{1000}{\pi h t(2R+t)(\rho)(C)}$; h = 120 in., R = 12 in., ρ = density, C = cost ($/lb).

c. Based on stress to produce 1% creep in 100 years (safety factor a = 0.5).

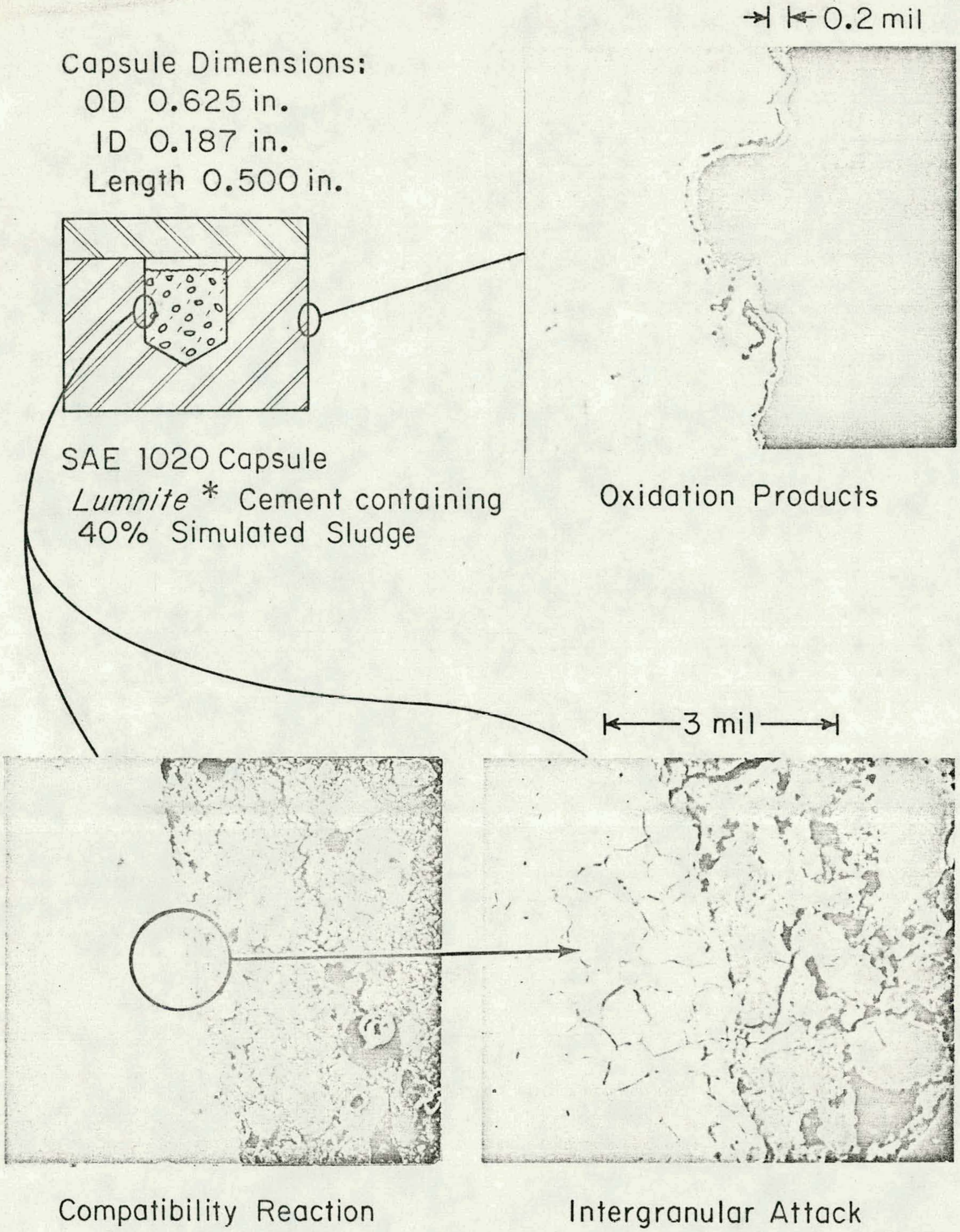
d. Based on stress to produce 1% creep (safety factor a = 1); rupture strength would not be exceeded.

e. Rupture strength would be exceeded.



10^5
10^4
10^3
500
Strength, psi
1020 CS
304 SS
Ultimate Tensile Strength
Rupture
1% Creep
100 yr Storage
Loss of Cooling
Fire
Glass, Continuous Melting
Glass, In-Can Melting
10
20
30
40
50
60
70
80
Larsen-Miller Parameter, P=T(18+log t) x 10^-3; T, °R; t, hr

Fig 1



*Trademark of Universal Atlas Division of US Steel for High-Alumina Cement.

Fig-2b

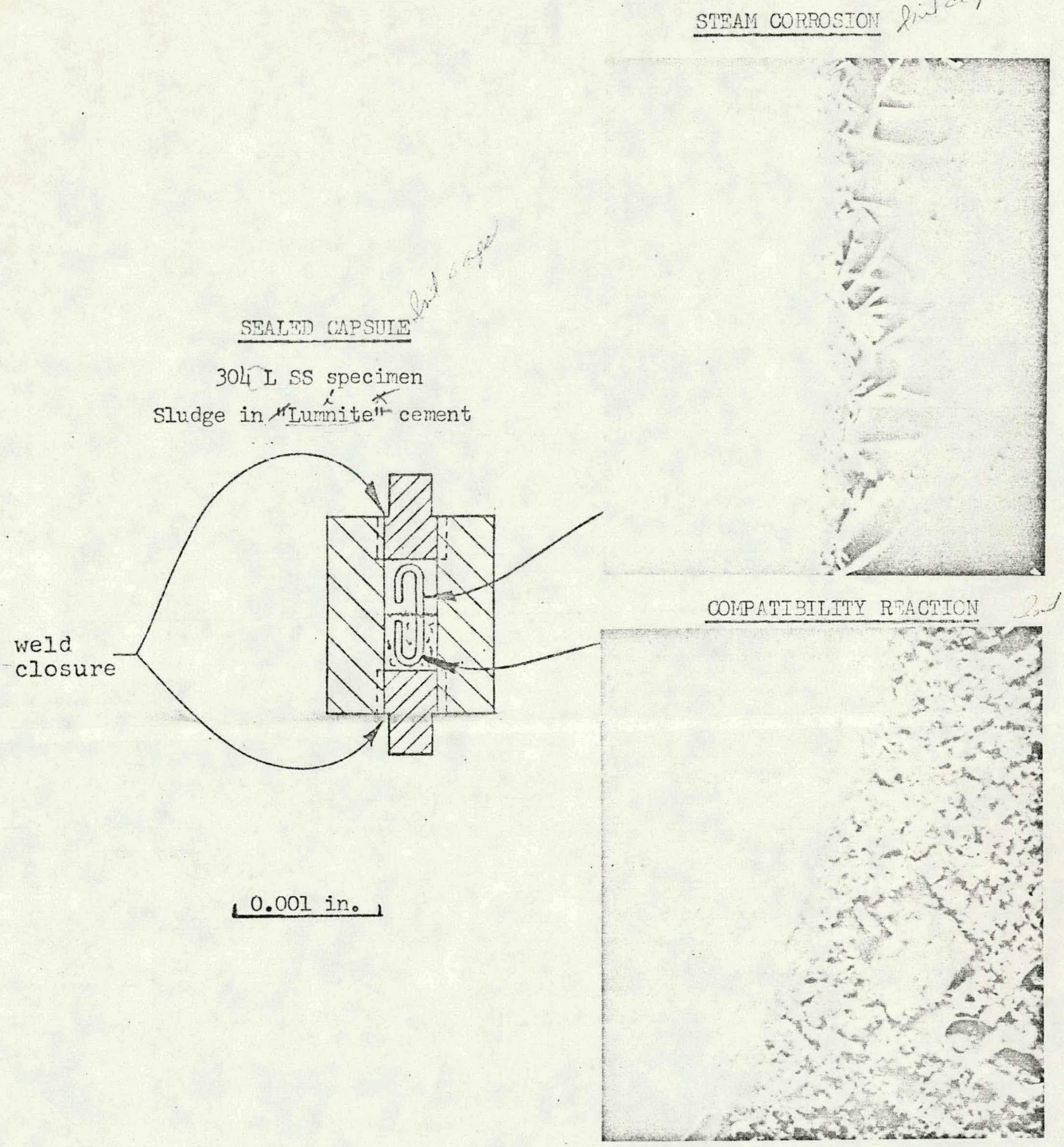


FIGURE 3 – TYPICAL SEALED CAPSULE HEATED 1000 HR AT 300 C.

* Trademark of Universal Atlas Cement Company.

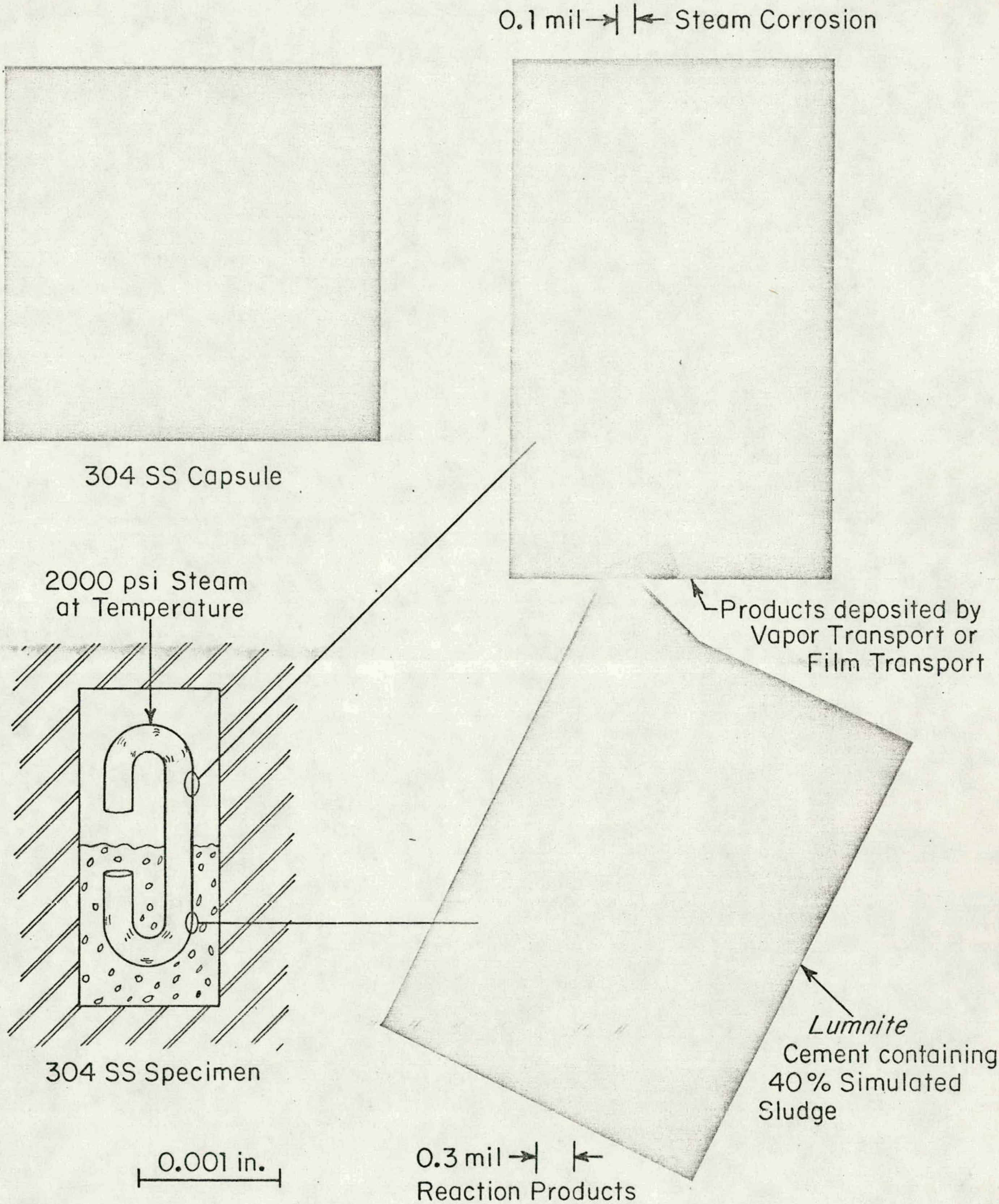


Fig. 3

~~FIG. 28 PG 79~~

~~85% Ky. 28~~

~~DP-76-1-7~~

304L SS GLASS MELTER

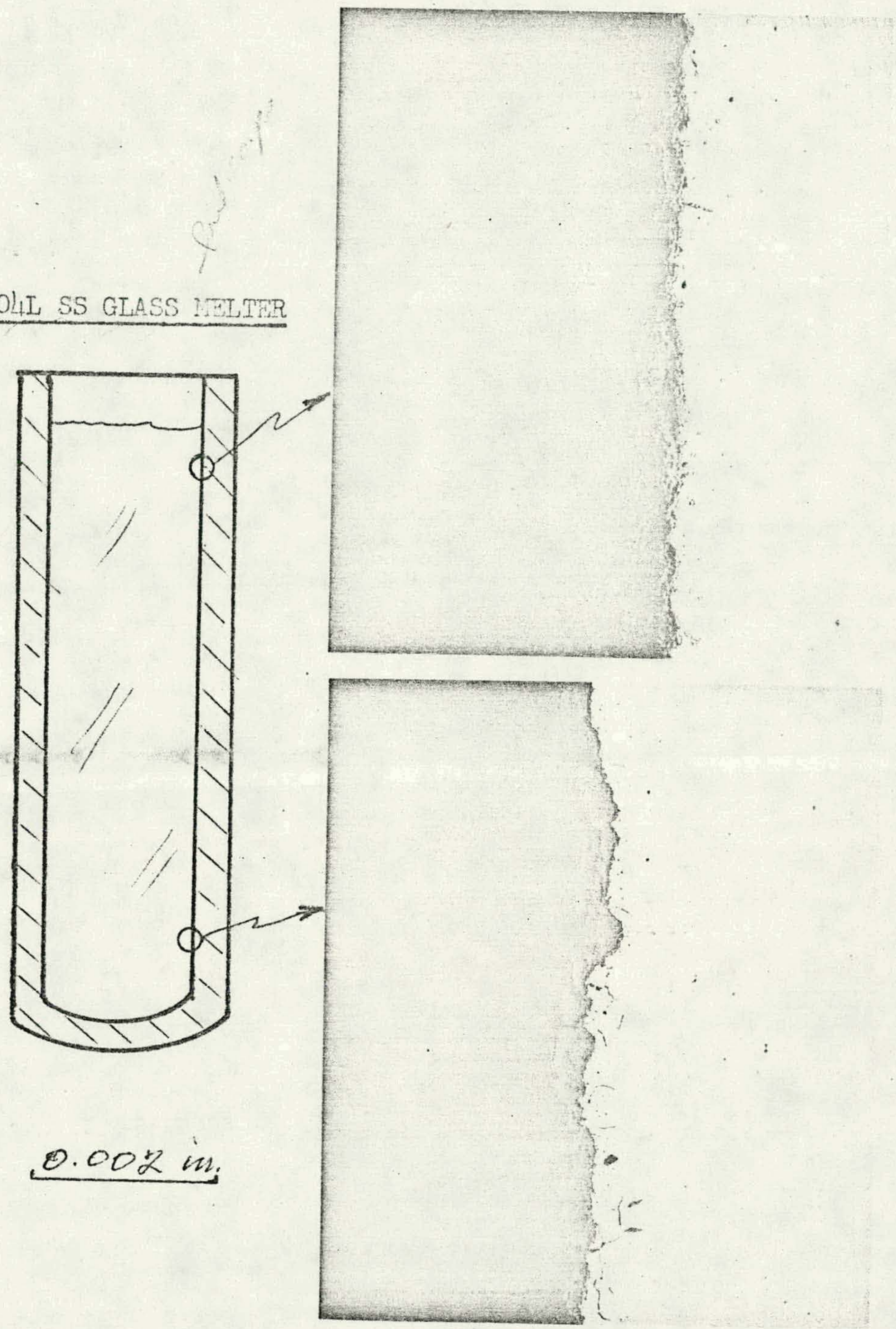


FIGURE 4 - REACTIONS DURING IN-CAN MELTING OF GLASS-

OP-MS-76-66
FIG 4
609. K-77

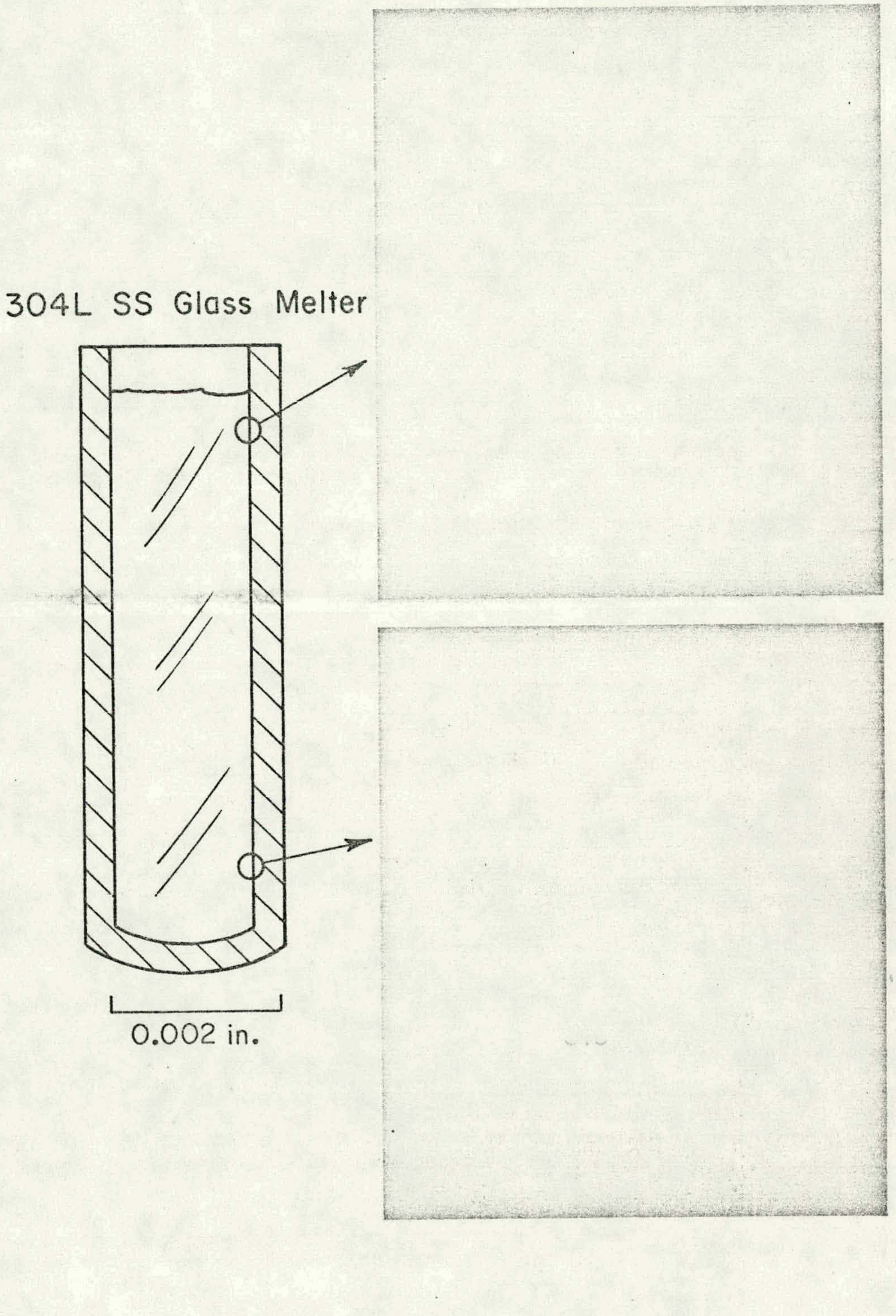
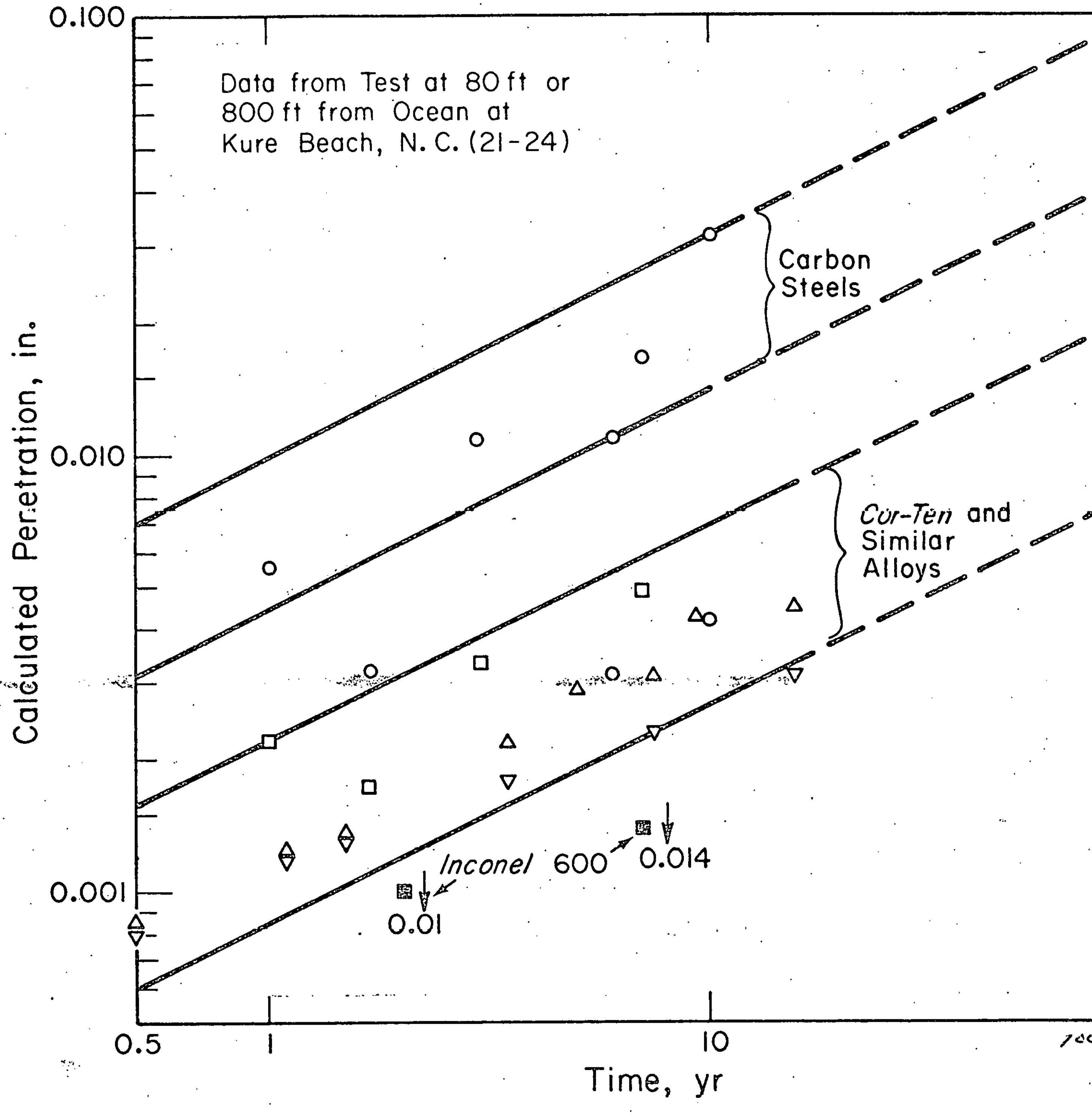


Fig. 4



Data from Test at 80 ft or
800 ft from Ocean at
Kure Beach, N. C. (21-24)
Carbon
Steels
Cor-Ten and
Similar
Alloys
Inconel 600
0.014
0.01
0.100
0.010
0.001
Calculated Penetration, in.
0.5
1
10
Time, yr

FIG 5 60%

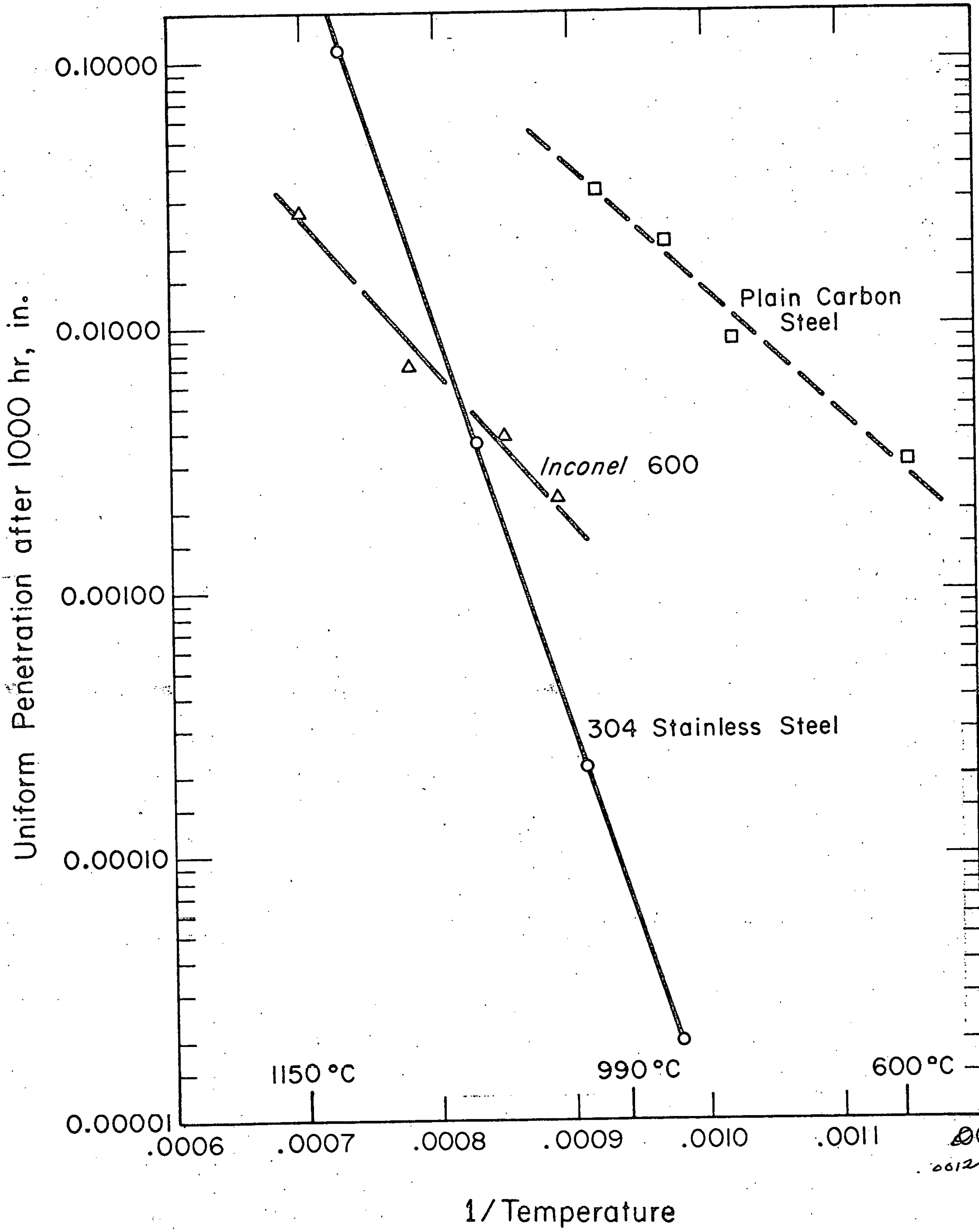

Uniform Penetration after 1000 hr, in.
0.10000
0.01000
0.00100
0.00010
0.00001
Plain Carbon Steel
Inconel 600
304 Stainless Steel
1150 °C
990 °C
600 °C
.0006
.0007
.0008
.0009
.0010
.0011
1/Temperature

Fig. 6

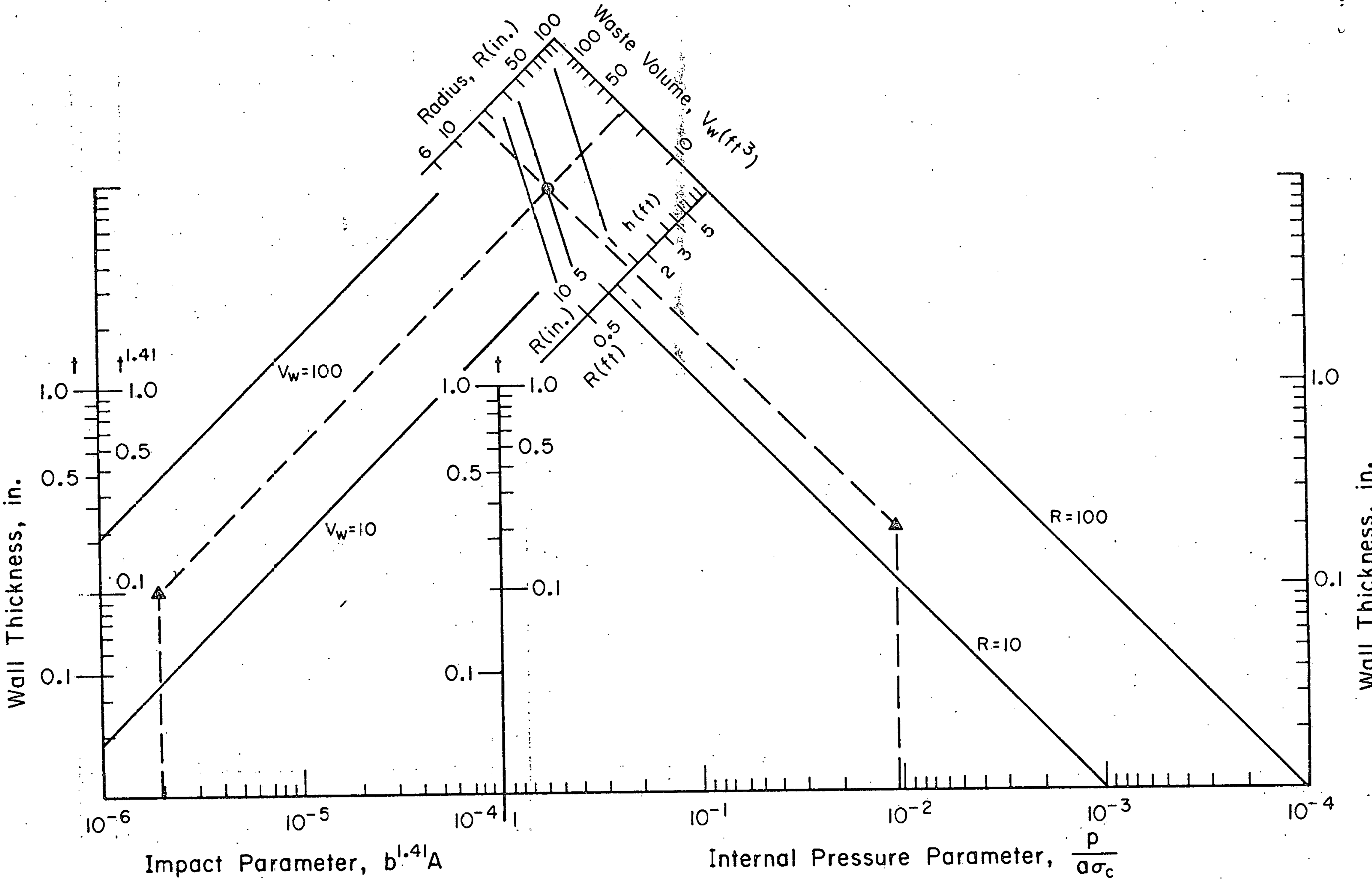

Radius, R(in.)
6
10
50
100
Waste Volume, $V_w(ft^3)$
100
50
10
h(ft)
2
3
5
10
5
R(in.)
0.5
R(ft)
V_w=100
V_w=10
R=100
R=10
t
$t^{1.41}$
1.0
0.5
0.1
Wall Thickness, in.
10^{-6}
10^{-5}
10^{-4}
1
10^{-1}
10^{-2}
10^{-3}
10^{-4}
Impact Parameter, $b^{1.41}A$
Internal Pressure Parameter, $\frac{p}{a\sigma_c}$

Fig. 7

FIGURE 1 - Typical Larsen-Miller plots.

FIGURE 2 - Typical compatibility capsule heated 10,000 hours at 350 C.

FIGURE 3 - Typical sealed capsule heated 1000 hours at 300 C.

FIGURE 4 - Reactions during "in-can" melting of glass.

FIGURE 5 - Atmospheric corrosion of candidate alloys.

FIGURE 6 - High-temperature oxidation of candidate alloys.

FIGURE 7 - Effect of materials properties on container dimensions.

For 100 year storage of concrete in a 1020 carbon steel container and safety factors of b = 1.5 and a = 0.5, the Impact Parameter = 1.95×10^{-6} and the Pressure Parameter = 1.25×10^{-2}. Entering the graph with these values on the abscissa, extrapolation from a wall thickness of 0.2 in. gives container dimensions of 30 ft^3 volume, 16.7 in. radius, and 5.2 feet height.