

Survey of Degradation Modes of Candidate Materials for High-Level Radioactive-Waste Disposal Containers

Overview

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Contents

List of Volumes of the Survey	iv
Acronyms	v
Abstract	1
Introduction	2
1. Phase Stability	3
2. Oxidation and Corrosion	4
3. Localized Corrosion and Stress Corrosion Cracking of Austenitic Alloys	10
4. Stress Corrosion Cracking of Copper-Based Alloys	12
5. Localized Corrosion of Copper-Based Alloys	18
6. Effects of Hydrogen in Austenitic and Copper-Based Alloys	20
7. Weldability of Austenitic Alloys	20
8. Weldability of Copper-Based Alloys	22
Conclusions and Future Work	23
Acknowledgments	24
References	25
Appendix: Methodology of the Literature Search	27

List of Volumes of the Survey

This overview summarizes the eight-volume report *Survey of Degradation Modes of Candidate Materials for High-Level Radioactive-Waste Disposal Containers*. Sections 1 through 8 of this overview summarize Volumes 1 through 8 of the survey, respectively. The titles of all of the volumes of the survey are as follows:

Overview

Volume 1: Phase Stability

Volume 2: Oxidation and Corrosion

Volume 3: Localized Corrosion and Stress Corrosion Cracking of Austenitic Alloys

Volume 4: Stress Corrosion Cracking of Copper-Based Alloys

Volume 5: Localized Corrosion of Copper-Based Alloys

Volume 6: Effects of Hydrogen in Austenitic and Copper-Based Alloys

Volume 7: Weldability of Austenitic Alloys

Volume 8: Weldability of Copper-Based Alloys

Acronyms

bcc	body-centered cubic
CDA	Copper Development Association
fcc	face-centered cubic
HAZ	heat-affected zone
IGSCC	intergranular stress corrosion cracking
LC	localized corrosion
MIC	microbiologically induced corrosion
NNWSI	Nevada Nuclear Waste Storage Investigations Project
SCC	stress corrosion cracking
SCE	saturated calomel electrode
TGSCC	transgranular stress corrosion cracking
WZ	weld zone

Survey of Degradation Modes of Candidate Materials for High-Level Radioactive-Waste Disposal Containers

Overview

Abstract

Three iron- to nickel-based austenitic alloys and three copper-based alloys are being considered as candidate materials for the fabrication of high-level radioactive-waste disposal containers. The austenitic alloys are Types 304L and 316L stainless steels and the high-nickel material Alloy 825. The copper-based alloys are CDA 102 (oxygen-free copper), CDA 613 (Cu-7Al), and CDA 715 (Cu-30Ni).

Waste in the forms of both spent fuel assemblies from reactors and borosilicate glass will be sent to the prospective repository at Yucca Mountain, Nevada. The decay of radionuclides will result in the generation of substantial heat and gamma radiation. Container materials may undergo any of several modes of degradation in this environment, including undesirable phase transformations due to a lack of phase stability; atmospheric oxidation; general aqueous corrosion; pitting; crevice corrosion; intergranular stress corrosion cracking; and transgranular stress corrosion cracking. Problems specific to welds, such as hot cracking, may also occur.

A survey of the literature has been prepared as part of the process of selecting, from among the candidates, a material that is adequate for repository conditions. The modes of degradation are discussed in detail in the survey to determine which apply to the candidate alloys and the extent to which they may actually occur. The eight volumes of the survey are summarized in Secs. 1 through 8 of this overview. The conclusions drawn from the survey are also given in this overview.

The candidates are only tentatively ranked on the basis of the survey. It is important to note that the rankings in this report are based on information in the general technical literature, and while the literature includes a broad range of environmental conditions, the rankings are not based on experimental work specific to Yucca Mountain. To as practical an extent as possible, the literature was surveyed for and the results were applied to the physical, chemical, metallurgical, and mechanical conditions that are expected to occur in the case of the emplaced metal containers at the Yucca Mountain repository. The basis of the rankings for each degradation mode is discussed in detail in Secs. 1 through 8.

On the basis of the literature review, Alloy 825 and CDA 715 are believed to have superior overall properties for the conditions expected in Yucca Mountain. For example, considering the metal alone as the thermodynamic system, Alloy 825 has a thermodynamically stable austenitic structure, whereas Types 304L and 316L stainless steels are metastable. CDA 715 is a simple solid solution of nickel in copper and does not suffer from the problems of internal oxidation encountered with CDA 102. CDA 613 depends on iron precipitates for mechanical strength; these may grow in size over extended periods of time. Though pitting and crevice corrosion may be encountered with all of the candidates, Alloy 825 and CDA 715 seem the least susceptible to these forms of attack. Under conditions known to induce stress corrosion cracking (SCC) of

Types 304L and 316L stainless steels, Alloy 825 appears resistant to this mode of degradation. Similarly, CDA 715 is the only copper-based alloy that appears to have good resistance to SCC over a broad range of conditions known to cause failure in CDA 102 and CDA 613.

This ranking of the candidate alloys is only tentative. The selection of an alloy will be made prior to design studies and will be based on formal selection criteria, which include other factors besides corrosion properties. The selection criteria are being formulated by the Metal Barrier Selection and Testing Task and are being peer-reviewed by eminent members of the scientific and engineering professions.

Introduction

Iron-based and nickel-based austenitic alloys as well as copper-based alloys are being considered as candidate materials for fabrication of high-level radioactive-waste disposal containers. The austenitic materials are Types 304L and 316L stainless steels and Alloy 825 (Incoloy 825). The copper-based alloys are CDA 102 (oxygen-free copper), CDA 613 (Cu-7Al), and CDA 715 (Cu-30Ni). These materials are leading candidates because of their good corrosion properties, their wide use in the marine, nuclear, and process industries, and their reasonable costs.

Waste in the forms of both spent fuel assemblies from reactors and borosilicate glass will be sent to the prospective repository at Yucca Mountain, Nevada, for disposal. The containers must be retrievable for 50 yr and exhibit substantially complete containment for 300 to 1000 yr following repository closure [1, 2]. (Retrieval entails removing the container with waste from the repository, possibly for reprocessing spent fuel.) Shortly after emplacement of the containers in the repository, radioactive decay of the stored waste will result in substantial heat transfer to the surroundings and in emission of gamma radiation. For waste packages with the highest thermal outputs, the surface temperature of the containers will rise to a peak of about 250°C. After 100 yr, the temperature will drop to about 150°C and will continue dropping slowly. Containers with lower heat output and those located at the edge of the repository will develop lower temperature peaks.

Container materials could undergo any of several modes of degradation after emplacement in this environment, including atmospheric oxidation, general aqueous corrosion, and various forms of localized corrosion (LC) and stress corrosion cracking (SCC). LC phenomena include crevice corrosion and pitting. Intergranular stress

corrosion cracking (IGSCC) and transgranular stress corrosion cracking (TGSCC) are forms of stress corrosion. In many alloy systems, LC and stress corrosion are interrelated because sites of LC attack become sources of initiation of stress corrosion. Corrosion attack can be enhanced by the growth of microorganisms that may be introduced into the repository or that may be present there naturally (autochthonous), as these could significantly alter the chemical conditions.

Other modes of degradation must also be considered. The effects of hydrogen share some common features with SCC in many metals and alloys. Phase instability can result in the formation of brittle structures in some of the candidate alloys. Welded regions and heat-affected zones (HAZs) of welds often show enhanced corrosion attack or susceptibility to embrittlement because of changes in metallurgical composition and residual stresses occurring in the welds.

The Metal Barrier Selection and Testing Task of the Nevada Nuclear Waste Storage Investigations (NNWSI) Project has to select a candidate material that is adequate for repository conditions. This selection will be based on a survey of the literature, corrosion testing by Lawrence Livermore National Laboratory and subcontractors, predictions from modeling, and a comparison of costs. The literature on the modes of degradation of the candidate materials is surveyed in detail in the report *Survey of Degradation Modes of Candidate Materials for High-Level Radioactive-Waste Disposal Containers*. This survey, of which the present report is an overview, is a critical analysis of the relevant technical literature on the various modes of degradation. The eight volumes of the survey are summarized in Secs. 1 through 8 of this overview, respectively. Over 1000 individual articles are cited in the survey. To

as practical an extent as possible, the articles were surveyed for and the results were applied to the physical, chemical, metallurgical, and mechanical conditions that are expected to occur in the case of the emplaced metal containers at the Yucca Mountain repository. The methodology and extent of the literature search are discussed in the Appendix.

The candidate alloys are tentatively ranked according to their resistance to each mode of

degradation. This ranking will play an important part in the selection of the metal barrier material for future design work.

Section 8 of this overview is followed by a section which presents the conclusions drawn from the survey and which gives an outline of future work that is derived from these conclusions.

1. Phase Stability

In Volume 1 of the survey of degradation modes [3], Bullen and Gdowski have surveyed the literature (75 articles) on the phase stability of the metal-barrier candidate materials. They found that both Types 304L and 316L stainless steels are metastable because of the possible coexistence of body-centered cubic (α , bcc) and face-centered cubic (γ , fcc) phases. In contrast, the austenite phase in Alloy 825 is thermodynamically stable when the metal itself constitutes the thermodynamic system under consideration, as illustrated in the ternary phase diagram shown in Fig. 1. Note that this figure is for 650°C. As the temperature is lowered, both Types 304L and 316L stainless steels move from the austenite region through a martensite-plus-austenite region to a martensite region. Furthermore, martensitic transformations have been observed in Types 304L and 316L stainless steels, but no such transformations have been observed in Alloy 825. An intermetallic phase, σ , has been observed in Type 316L which can result in a degradation of mechanical properties. The formation of intermetallic phases has not been documented for Alloy 825. Furthermore, the authors found that the σ phase is promoted by the presence of elements such as silicon.

On the basis of their relative phase stabilities, the austenitic alloys are tentatively ranked as follows:

Alloy 825 (best) > Type 316L > Type 304L (worst)

The IGSCC of austenitic alloys occurs when these materials are subjected to a sufficiently severe combination of stress, corrosive environment, and sensitization, a term denoting increased susceptibility to attack following a thermal exposure that causes chromium-rich $M_{23}C_6$ carbides to precipitate at grain boundaries. Chromium carbide precipitation in stainless steels

occurs in the temperature range of 500 to 850°C (930 to 1560°F), with the rate of precipitation controlled by the diffusion of chromium. Precipitation results in the formation of a narrow chromium-depleted zone, adjacent to the carbides, in which the local chromium concentration is below 12 wt%. Since chromium is the element responsible for the formation of stable passive films in stainless steels, localized depletion of this element results in the establishment of an "active path" into the bulk material. $M_{23}C_6$ particles have been observed in all of the austenitic candidates: Types 304L and 316L stainless steels and Alloy 825. Control of carbon content in Type 304L or 316L to less than 0.035% is effective in minimizing the sensitization. Moreover, the problem is effectively eliminated in Alloy 825 by the addition of titanium. The titanium promotes the formation of titanium carbides, thus minimizing the loss of chromium.

Of the copper-based candidate alloys, it is obvious that oxygen-free copper (CDA 102) has the best phase stability. It is a single-phase material with an fcc crystal structure. In the case of CDA 613, aluminum is completely soluble in the copper matrix over practical ranges of temperature. However, iron precipitates are present and are responsible for the strength and hardness of this alloy. The phase diagram for copper-aluminum is shown in Fig. 2. The cupronickel alloy CDA 715 exists as a 30% solid solution of nickel in copper. A two-phase region, $\alpha_1 + \alpha_2$, has been predicted by thermodynamic calculations at lower temperatures but has not been observed. These features are shown in the binary phase diagram in Fig. 3. A small quantity of manganese (0.05%) is present in CDA 715 and serves as a deoxidant. Note that manganese is soluble in the matrix. Iron (0.05%) enhances the corrosion resistance of CDA 715 and is insoluble in the matrix. However, iron precipitation is

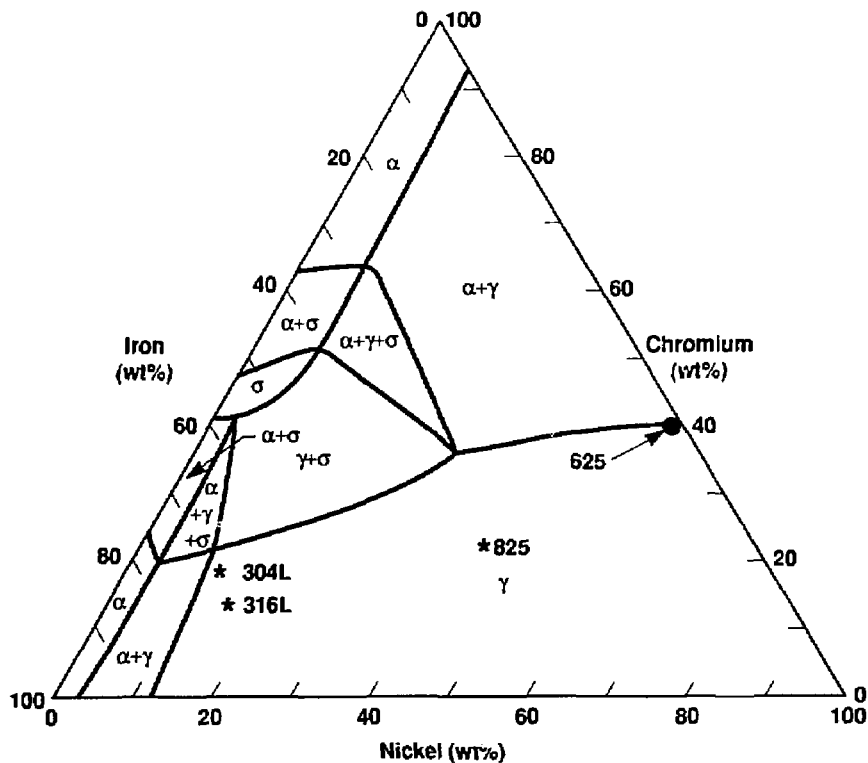


Figure 1. Iron-chromium-nickel equilibrium phase diagram at 650°C [3]. Alloy 825 is thermodynamically stable. Types 304L and 316L stainless steels are close to two-phase regions and are known to be metastable.

prevented by the slow rate of diffusion of iron in the matrix. Precipitation over long periods of time could result in a degradation of the mechanical properties.

On the basis of their relative phase stabilities, the copper-based alloys are tentatively ranked as follows:

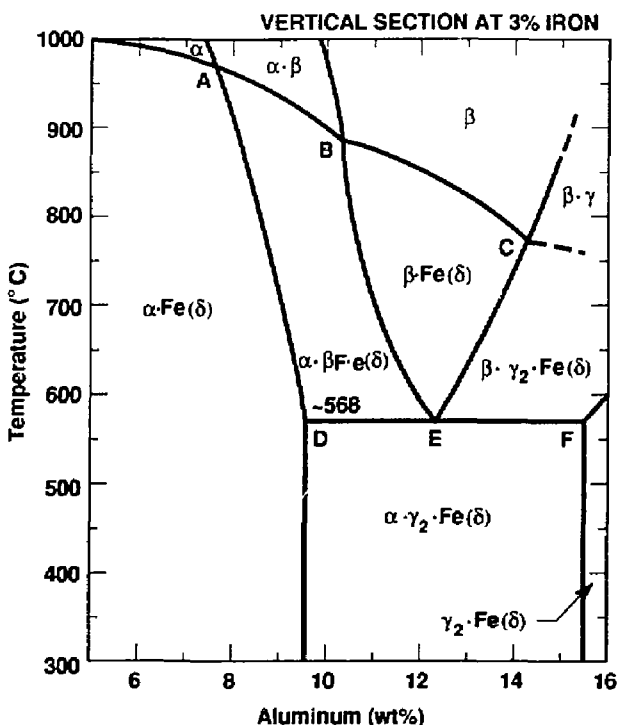
CDA 102 (best) > CDA 715 = CDA 613 (worst)

2. Oxidation and Corrosion

In Volume 2 of the survey of degradation modes [7], Gdowski and Bullen have reviewed the literature (104 articles) on the general corrosion and oxidation of the six candidate alloys. They found that the excellent resistance of austenitic alloys to corrosion is due to their capacity to become passivated with a film of chromium oxide that is 10 to 50 Å thick. At least 12 wt% chromium is necessary for the formation of a protective Cr_2O_3 film. Molybdenum is introduced

into the austenitic candidates for stabilization of the passive films.

Figure 4 shows representative data on corrosion rates of the austenitic alloys—Types 304 and 316 stainless steels and Alloy 825—in rural, urban, and industrial atmospheres [7]. Rates were calculated from weight-loss measurements of coupons after environmental exposure at Bayonne and Newark, New Jersey; Kure Beach, North Carolina; Bridgeport, Connecticut; Point Reyes,



	Point					
	A	B	C	D	E	F
°C	970	885	770	-568	-568	-568
Al (%)	7.6	10.3	~14.3	9.5	12.3	15.5

Figure 2. Copper-aluminum equilibrium phase diagram at 3% iron [3, 4].

California; State College, Pennsylvania; the Panama Canal Zone; Sheffield, England; and places throughout Canada. Findings on the atmospheric oxidation of austenitic candidates indicate the following tentative ranking:

Type 316 (best) > Alloy 825 > Type 304 (worst)

Figure 5 shows representative data on corrosion rates of Types 304 and 316 stainless steels and Alloy 825 in seawater (Panama Canal) and in superheated steam (566 and 621°C) [7]. The data are also presented in Table 1. These findings indicate a slightly different tentative ranking:

Alloy 825 (best) > Type 316 > Type 304 (worst)

The lifetimes of containers fabricated from the austenitic alloys would not be limited by uniform atmospheric oxidation and corrosion. Furthermore, the lifetimes of containers fabricated from either Type 316 (similar to Type 316L) or Alloy 825 would not be limited by uniform corrosion in seawater or steam. Such aqueous corrosion might, however, limit the lifetimes of containers fabricated from Type 304 (similar to Type 304L).

McCright et al. [8] conducted tests and found that all of the austenitic alloys perform comparably in water at 50, 80, and 100°C as well as in steam at 100 and 150°C. These data are illustrated in Fig. 6. The results also indicate that the lifetimes of containers fabricated from the austenitic

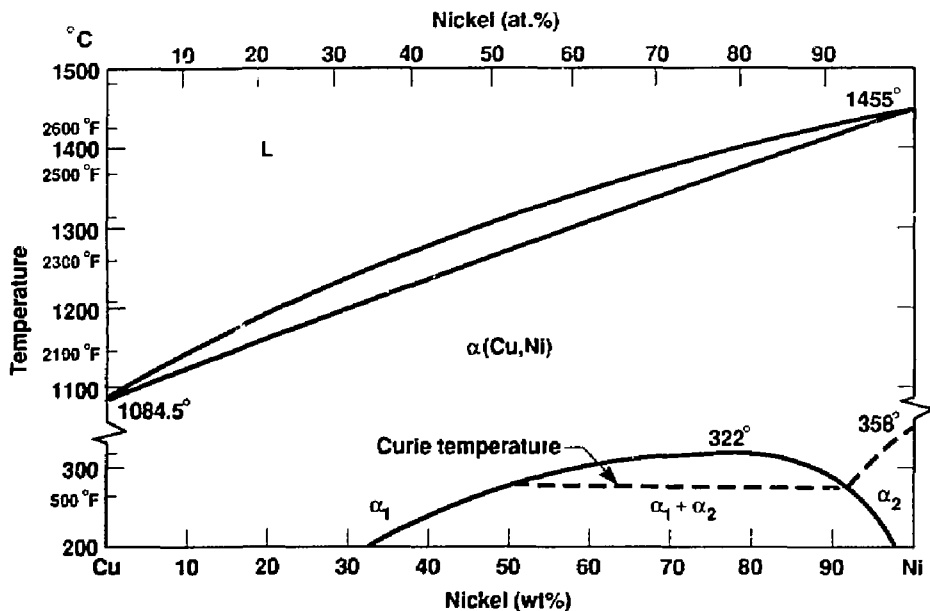


Figure 3. Copper-nickel equilibrium phase diagram [3, 5, 6]. CDA 715 is a single-phase alloy. Note that Cu-30Ni is a solid solution of nickel in copper.

Table 1. U-bend specimens after 1 month in 0.8% NaCl and 0.5% CH₃COOH at 141°C [14].

Alloy	Corrosion rate (mm/yr)	Localized corrosion	SCC
304	0.03	Severe pitting	Yes
304L	0.02	Severe pitting	Yes
316	0.01	Severe pitting	Yes
316L	<0.01	Pitting	Yes ^a
825	<0.01	No attack	No

Note: SCC was observed in the 300-series alloys after 30 days of exposure; no SCC was observed in Alloy 825 or in the alloys with higher nickel content.

^a One specimen out of two cracked.

candidates would not be threatened by general corrosion and oxidation in hot water and steam.

The literature on the three copper-based alloys, CDA 102, CDA 613, and CDA 715, was also reviewed. In comparing the copper alloys, it is necessary to evaluate the structure of the oxide films. Formation of stable, thin oxide films on metals results in a diffusion barrier that inhibits further oxidation. Thus, while oxidation rates are initially linear with time, as the film is formed the rates decrease to a parabolic or other similar form.

It was found that the oxide that forms on CDA 102 in air at temperatures below 200°C is primarily Cu₂O, whereas a combination of Cu₂O and CuO forms at higher temperatures. The ratio of CuO to Cu₂O depends upon the partial pressure of oxygen. The growth of this film is limited by the solid-state transport of cuprous cations through the oxide. Consequently, growth of oxide would be expected to obey a parabolic rate law. It was also found that oxide films that form on aluminum bronze (CDA 613) and cupronickel

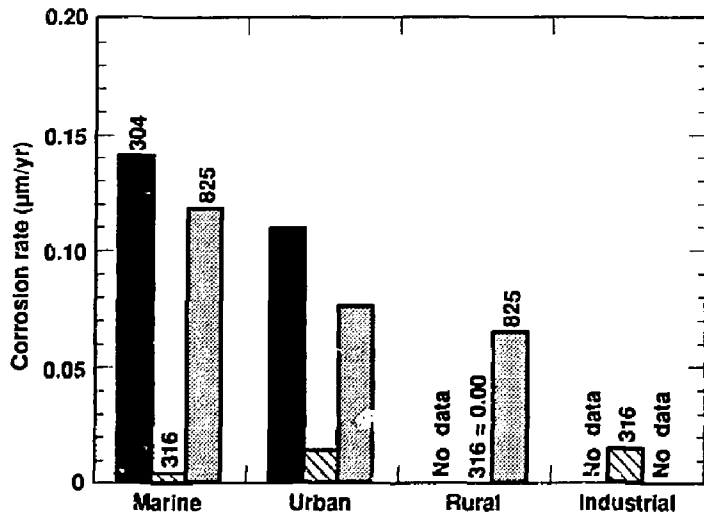


Figure 4. Atmospheric corrosion rates of the austenitic alloys [7]. Container lifetime is not limited by the atmospheric corrosion of austenitic alloys.

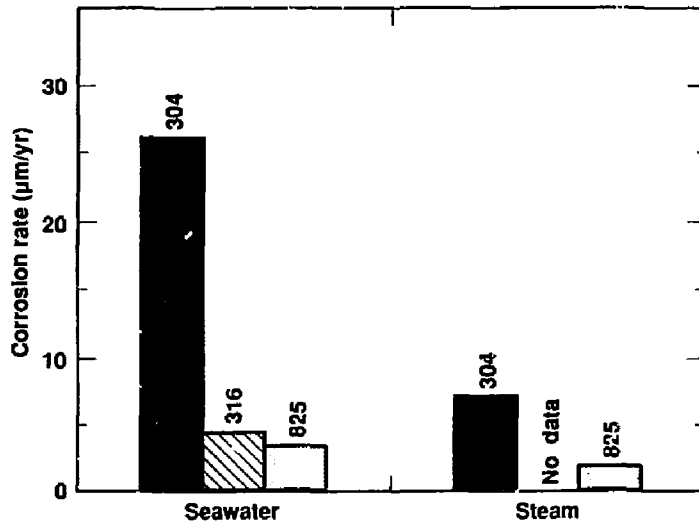


Figure 5. Aqueous corrosion rates of the austenitic alloys [7]. Container lifetime in the case of Type 304 (similar to 304L) may be limited by aqueous corrosion in salt solution (seawater).

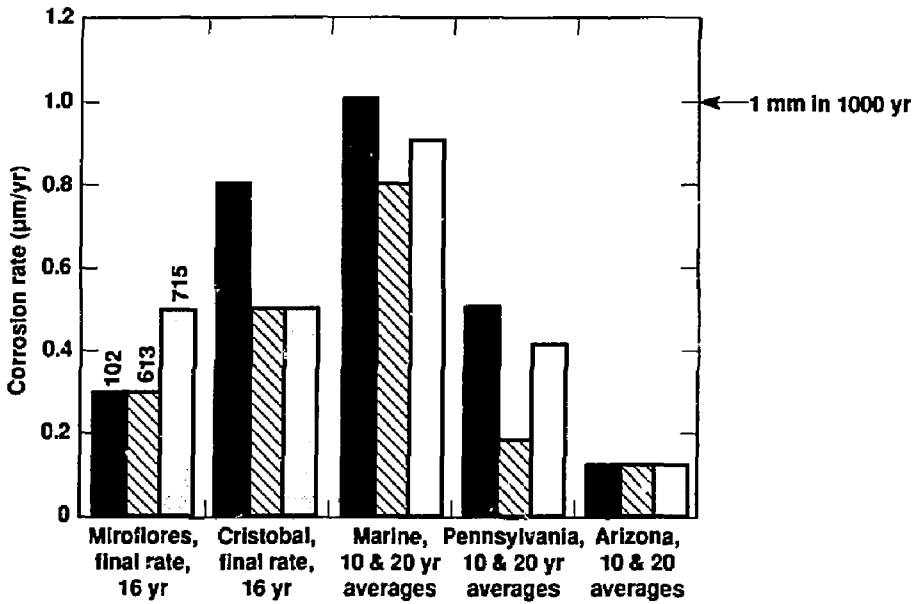


Figure 6. Atmospheric corrosion rates of the copper-based alloys [7]. Container lifetime is not limited by the atmospheric corrosion of copper-based alloys.

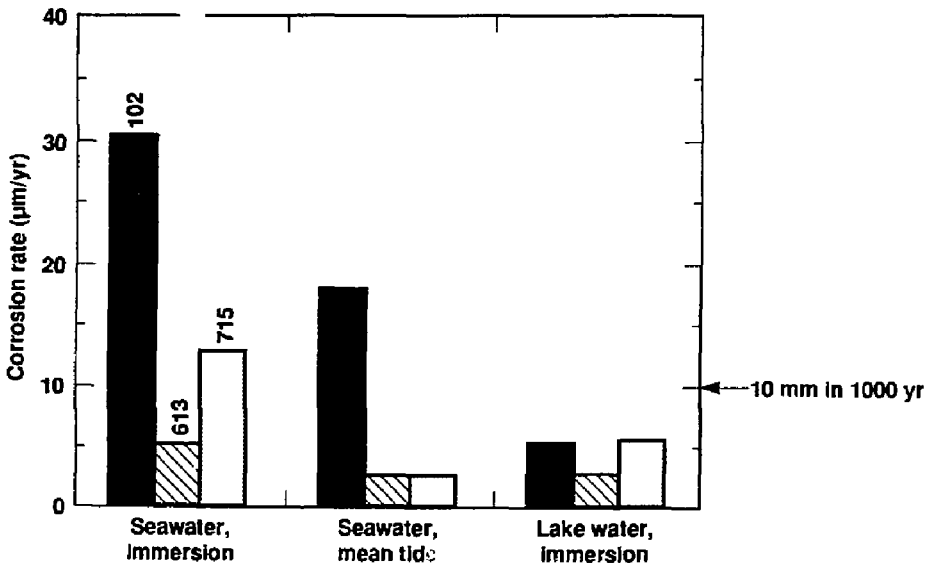


Figure 7. Aqueous corrosion rates of the copper-based alloys [7]. The higher aqueous corrosion rates of CDA 102 and CDA 715 in salt solutions are noteworthy. CDA 613 has the best performance because of a passive film of alumina.

(CDA 715) grow more slowly than the film that forms on oxygen-free copper (CDA 102). The oxide film that forms on CDA 613 in air is primarily Al_2O_3 . This alumina film serves as a diffusion barrier against the solid-state transport of cuprous cations, thereby limiting the growth of CuO and Cu_2O . In contrast, the film that forms on CDA 715 in air is primarily Cu_2O or CuO , with an interlayer of NiO between the copper oxide and the alloy substrate. The NiO interlayer also serves as a diffusion barrier to the migration of cuprous cations, thus tending to inhibit the rate of CuO and Cu_2O formation. It is not quite as effective, however, as the film of CDA 613.

Data for atmospheric corrosion rates of the copper-based alloys are shown in Fig. 7. It is seen that the lifetimes of containers fabricated from any of the copper-based candidate alloys would not be limited by atmospheric oxidation. Figure 8 shows data for the aqueous corrosion of these materials. While it is unlikely that container lifetime will be limited by aqueous corrosion, the higher corrosion rates of CDA 102 and 715 in concentrated salt solutions such as seawater are noteworthy. CDA 613 has the best resistance to uniform attack in concentrated salt solutions.

Figure 9 shows the results of gamma-irradiation tests on the copper-based alloys in water at 95°C, in air nearly saturated with water vapor at 95°C, and in moist air at 150°C. Clearly, CDA 613 has the lowest rate of oxidation. Furthermore, the relatively higher rates found in CDA 102 or CDA 715 in air nearly saturated with water vapor at 95°C are noteworthy.

The primary problem encountered with copper is internal oxidation and the associated hydrogen attack of the oxidized microstructure. Phosphorus is frequently added to serve as a deoxidant, as in the case of CDA 122.

On the basis of this information, we tentatively rank the copper-based alloys in order of their resistance to atmospheric oxidation and aqueous general corrosion as follows:

CDA 613 (best) > CDA 715 > CDA 102 (worst)

Since it has been found that rates of penetration due to pitting and SCC are usually orders of magnitude greater than rates of attack due to general corrosion, it is assumed that container life will probably be limited by some form of localized attack. Atmospheric and aqueous corrosion

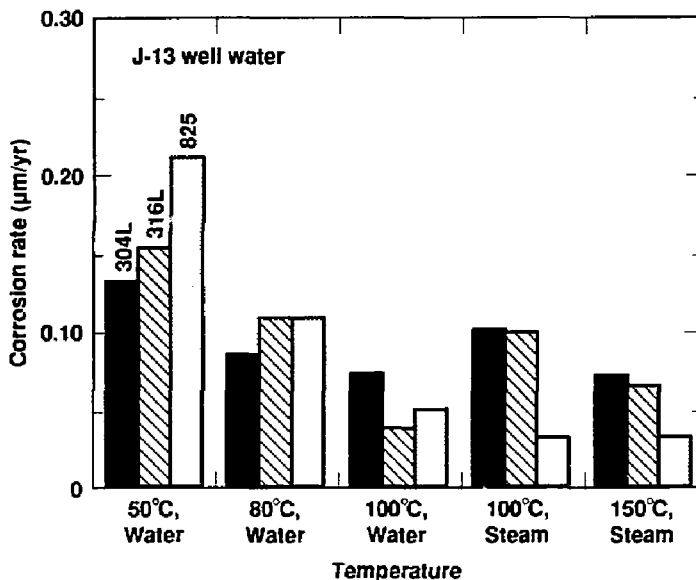


Figure 8. General corrosion and oxidation of austenitic alloys [8]. Container lifetime is not limited by these modes of degradation.

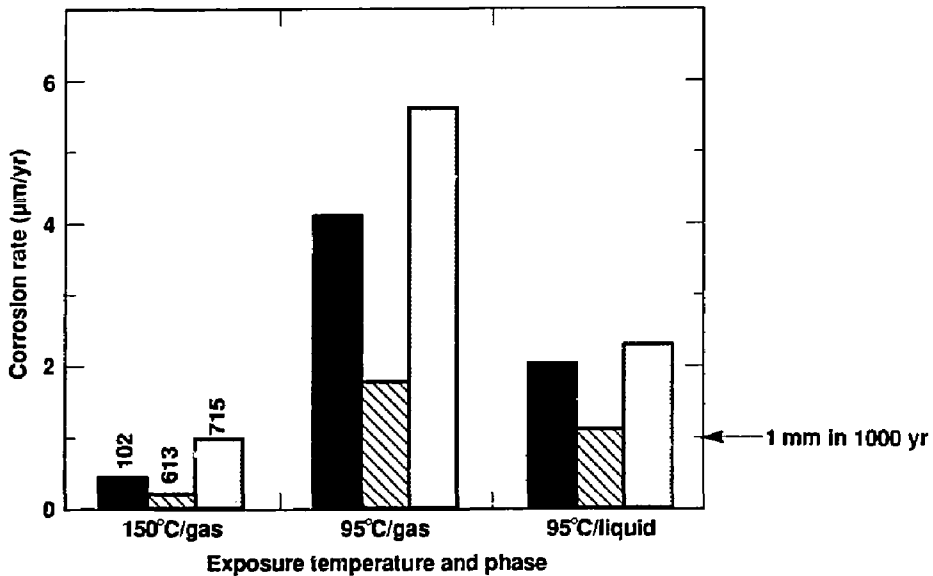


Figure 9. General corrosion rates for copper-based alloys under irradiation [9]. The relatively higher rates of corrosion for CDA 102 and CDA 715 in irradiated moist air at 95°C are noteworthy. CDA 613 has the best performance because of a passive film of alumina.

are assumed to be the least significant modes of degradation.

Atmospheric pollutants such as SO₂, H₂S, O₃, HCl, and Cl₂ enhance the rate of atmospheric oxidation of copper. Chemical species such as these would be detrimental to copper containers and

have not been considered in the tentative ranking given above. The rate of oxidation of copper appears to be insensitive to NH₃ and NO₂ even though these species are known to induce ICSCC and TGSCC.

3. Localized Corrosion and Stress Corrosion Cracking of Austenitic Alloys

Rates of penetration at local sites of corrosive attack are far more serious threats to container lifetime than rates due to general atmospheric and aqueous corrosion. In Vol. 3 of the degradation mode survey [10], Farmer et al. have surveyed the literature (211 articles) on LC and SCC of the three austenitic alloys.

First, all three austenitic candidates (Types 304L and 316L stainless steels and Alloy 825) have demonstrated pitting and crevice corrosion in chloride-containing environments. However, electrochemical polarization data indicate that Alloy 825 has greater resistance to pitting and crevice corrosion than Type 304L or 316L stainless

steel. The alloys are ranked as follows on the basis of their resistance to pitting and crevice corrosion:

Alloy 825 (best) > Type 316L > Type 304L (worst)

Resistance to pitting is quantified as the difference between the critical pitting potential, E_c , and the corrosion potential, E_{corr} . Large potential differences are indicative of materials having high resistances to pitting. In contrast, large differences between the repassivation potential, E_p , and E_c are indicative of materials having the greatest susceptibility to crevice

$$E_c = 2570 - 5.81 \cdot T + 0.07 \cdot T \cdot \text{pH} - 0.49 \cdot T \cdot \log[\text{Cl}^-] \text{ (mV, SCE)}$$

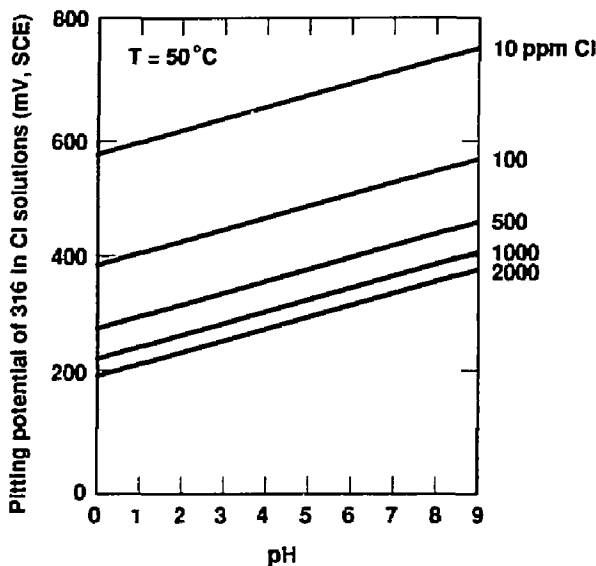


Figure 10. Variation of the pitting potential of Type 316 stainless steel in cellulose bleach solution at 50°C [11]. Increases in chloride concentration and decreases in pH increase the pitting susceptibility of austenitic alloys.

corrosion. As shown in Fig. 10, increases in chloride concentration and decreases in pH result in a decrease of the resistance to pitting of Type 316 stainless steel. Figures 11 and 12 compare data on resistances to pitting of Types 304 and 316 stainless steels and Alloys 825 and 625. Of the candidate alloys, Alloy 825 has the highest resistance to pitting. Data for Alloy 625 are included because this alloy can be used as a filler metal in welds of Alloy 825. Though Alloy 625 has good resistance to pitting, its phase stability may not be as good as that of Alloy 825.

Stressed austenitic specimens in aqueous environments containing sufficient dissolved chloride and oxygen undergo TGSCC. Combinations of concentrations of chloride and oxygen known to induce TGSCC are illustrated in Fig. 13. These data are for alkaline phosphate water, however. Since the repository will be closer to neutral pH and without phosphate, the data may not be relevant to the repository. Furthermore, in acidic chloride media, TGSCC has always been observed in U-bend specimens of Types 304L and 316L stainless steels. Under comparable circumstances,

however, TGSCC has not been reported for Alloy 825. These results are summarized in Tables 1 and 2. On the basis of their relative susceptibilities to SCC, the austenitic candidates are tentatively ranked as follows:

Alloy 825 (best) > Type 316L > Type 304L (worst)

Two special circumstances in the repository are also addressed in Vol. 3: the presence of gamma radiation, and microbiologically induced corrosion (MIC). Figure 14 shows that gamma irradiation enhances SCC of Type 304 stainless steel under some conditions. In contrast, Alloy 825 shows no change in its resistance to SCC in the presence of gamma irradiation. Though MIC and possible SCC have been observed for 300-series stainless steels, the nickel-based alloys such as Alloy 825 seem to be immune to microbiological problems.

From the survey on crevice corrosion, pitting, and SCC in austenitic alloys, it is found that Alloy 825 is the candidate most resistant to these forms of localized attack.

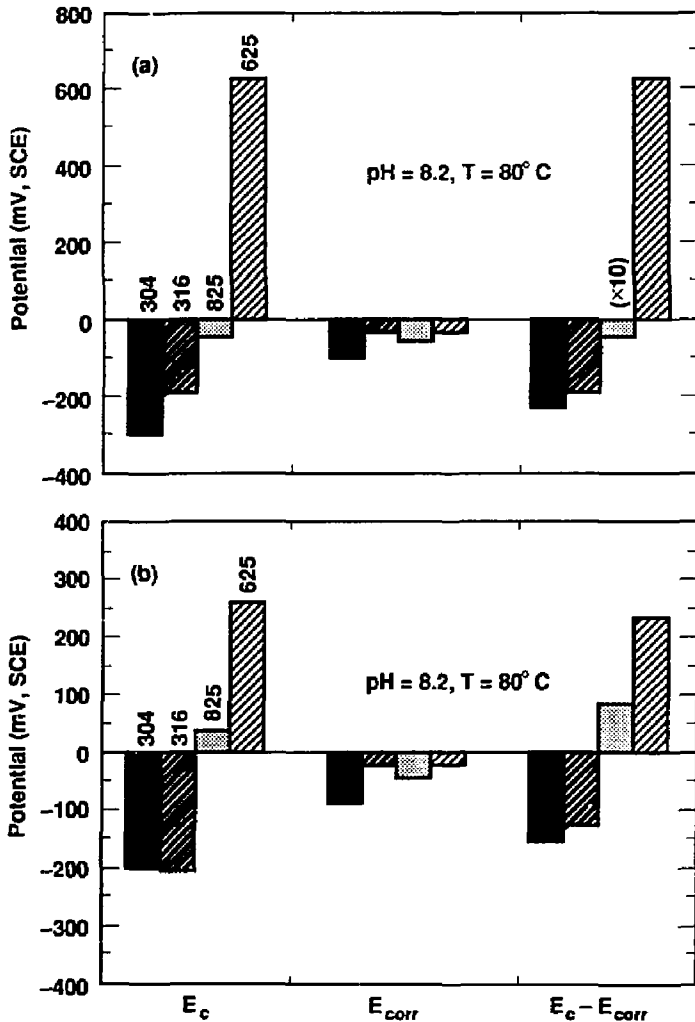


Figure 11. Pitting potential (E_c) and corrosion potential (E_{corr}) of candidate materials for (a) fast, and (b) slow scan tests in deaerated substitute ocean water [12]. Of the austenitic candidate alloys, Alloy 825 has the best resistance to pitting. Alloy 625 is a possible weld filler metal but is more expensive and has dubious phase stability.

4. Stress Corrosion Cracking of Copper-Based Alloys

In Volume 4 [16], Farmer et al. have surveyed the literature (117 articles) and established a tentative ranking of the copper-based alloys on the basis of their susceptibility to SCC. The copper-based alloys are susceptible to SCC in

moist, ammonia-containing environments as well as in nitrite solutions, cupric acetate solutions, and steam.

One scenario for the generation of SCC-promoting chemical species on the surfaces of the

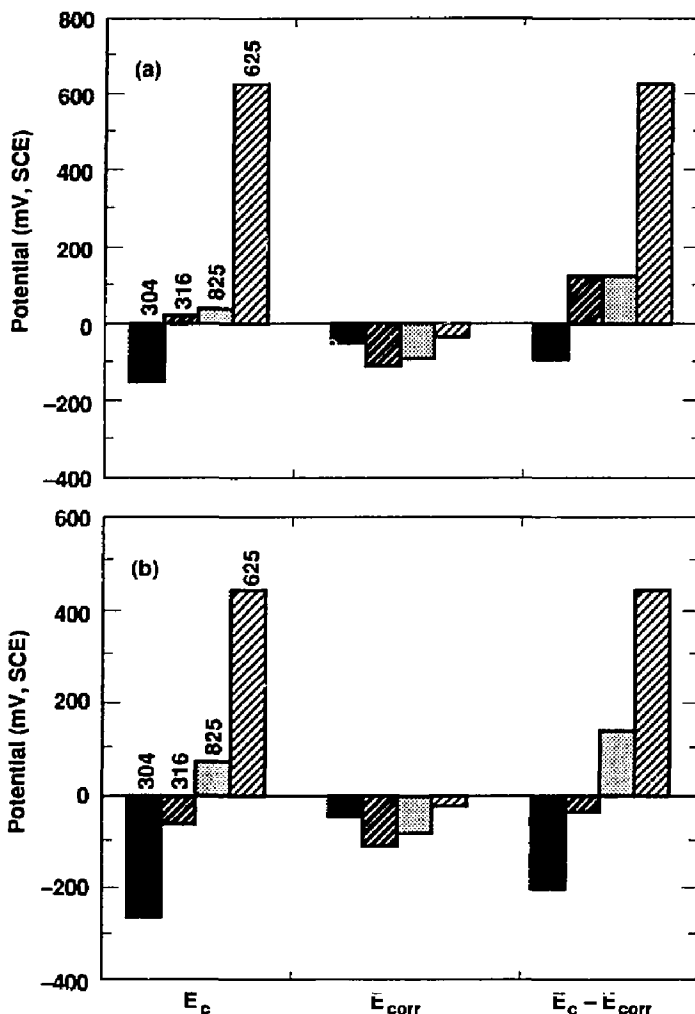


Figure 12. Same as Fig. 11, except that SO_2 -saturated seawater was used [12].

nuclear-waste containers is gamma radiolysis. For example, acidic nitrite could be generated in this manner. Radiation-induced SCC may therefore be possible. Figure 15 shows the dependence of the failure stress on nitrite concentration.

The generation of ammonia by radiolysis is unlikely, though a mechanism has been proposed in the literature for its generation by the cathodic reduction of nitrite to ammonia. This proposed mechanism was abandoned by the authors in subsequent publications. If ammonia did form, it would be oxidized by hydrogen peroxide.

Hydrogen peroxide is expected to be generated in moisture films but will undergo rapid catalytic decomposition on copper surfaces. There is a possibility of microbes in the repository generating a moist ammoniacal environment having nearly neutral pH. As illustrated in the Pourbaix (potential-pH) diagram shown in Fig. 16, a singularity exists at a pH of approximately 7.3 and a potential of approximately 275 mV, SCE. The passive film at the tip of SCC cracks is metastable under these conditions. Consequently, such environments promote the propagation of SCC. Fortunately, as shown in Fig. 17, the compositions

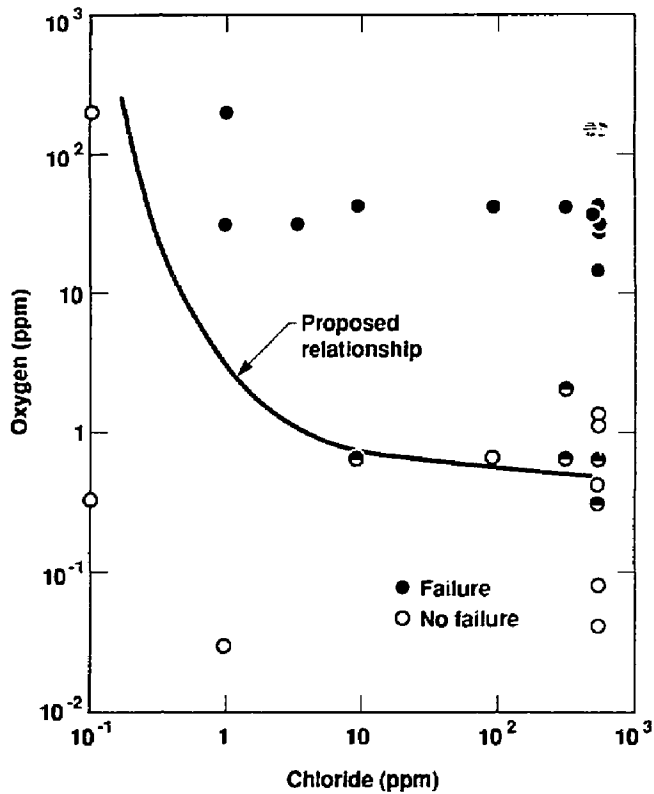


Figure 13. Proposed relationship between chloride and oxygen content of alkaline-phosphate-treated boiler water, and susceptibility to TGSCC of austenitic stainless steel exposed to the steam phase with intermittent wetting [13]. TGSCC would be expected in Type 304 stainless steel (in the unsensitized state similar to Type 304L) under repository conditions (1–10 ppm oxygen and 10–100 ppm chloride).

Table 2. U-bend specimens in 0.8% NaCl and HCl (pH 2.2) at 141°C [14].

Alloy	10-day exposure	30-day exposure
304	No cracking	Cracking
304L	No cracking	Cracking
316	No cracking	Cracking
316L	No cracking	Cracking
825	No cracking	No cracking

Note: All specimens with no cracking after the 10-day exposure were tested for an additional 20-day exposure. The temperature of the container was maintained at 285°C.

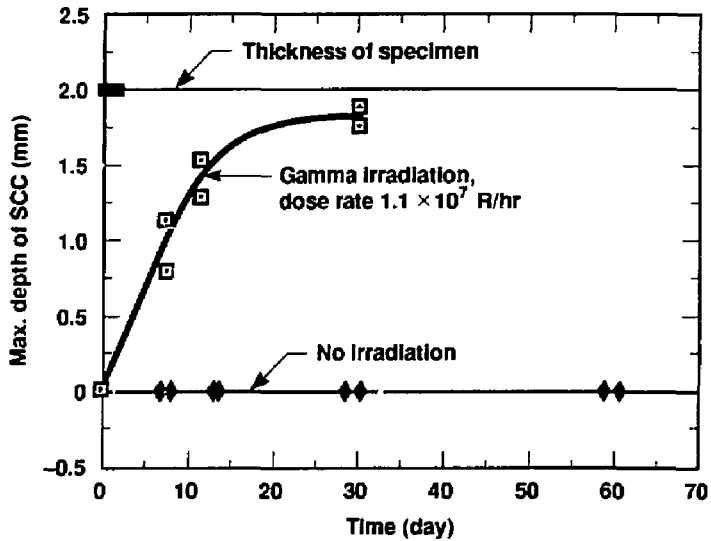


Figure 14. Effects of gamma irradiation on SCC failures of sensitized Type 304 stainless steel [15]. Gamma irradiation promotes IGSCC in sensitized microstructure.

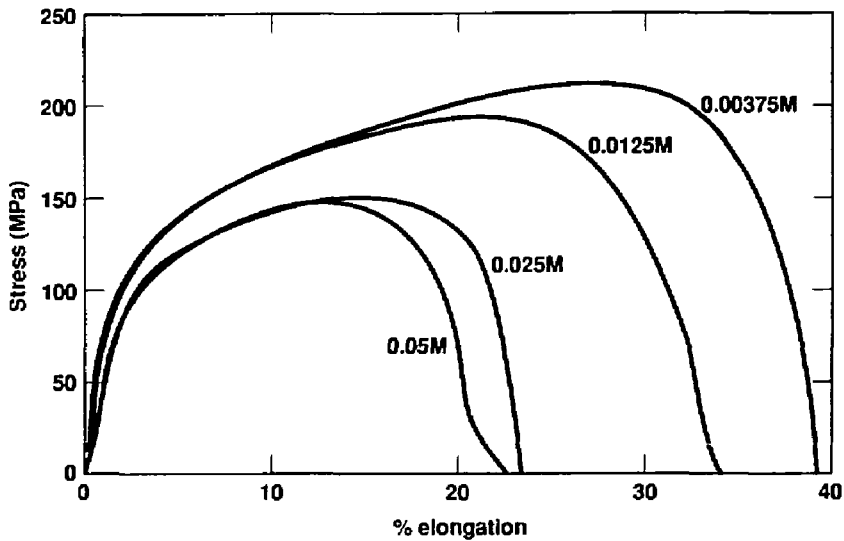


Figure 15. Stress-elongation curves obtained from slow strain rate tests of oxygen-free copper in aerated solutions of NaNO_2 at the concentrations shown [17]. Conditions were 25°C ; 100 mV, SCE ; and $2.6 \times 10^{-6}\text{ s}^{-1}$. Nitrite ions can be formed during radiolysis and can promote SCC of CDA 102.

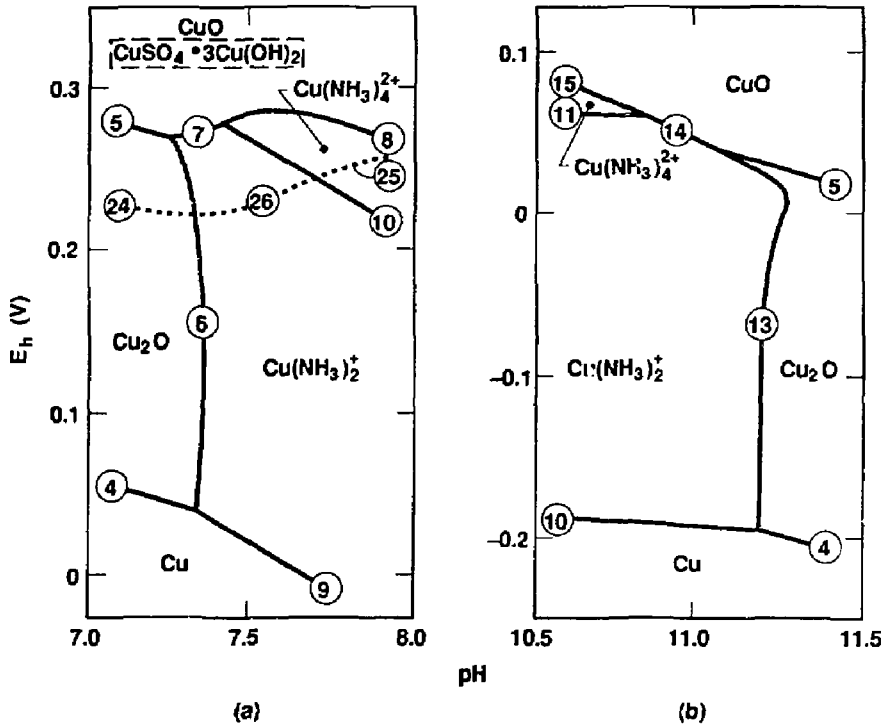


Figure 16. Detailed representations of the regions of singularity that occur on the Pourbaix (potential/pH) diagram around (a) pH 7.3, and (b) pH 11.3 [18]. Microbial growth can result in moist ammoniacal environments with a pH of approximately 7.3. Such environments are known to promote SCC in CDA 102 (as well as in the other copper-based alloys).

of CDA 613 (Cu-7Al) and CDA 715 (Cu-30Ni) have been optimized to minimize the susceptibility to SCC in moist ammoniacal environments.

More specifically, IGSCC of copper (CDA 102) is observed in nontarnishing ammoniacal solutions and cupric acetate solutions. TGSCC of copper (CDA 102) is observed in tarnishing ammoniacal and ammonium hydroxide solutions and nitrite solutions. Both film rupture and film-induced cleavage have been used to explain SCC of pure copper (CDA 102) and alpha brasses. The tarnish film involved in the SCC of CDA 102 is probably Cu_2O . Growth of this oxide obeys the parabolic law, which is consistent with the film-rupture model as well as the slip dissolution-repassivation model. In the $\text{NH}_3\text{-H}_2\text{O-Cu}$ system, the susceptibility of copper to SCC is greatest at combinations of potential and pH (7.2 or 11.3) where the passive film is metastable.

Note that the repository water will probably have a pH of about 7. Copper deoxidized with phosphorus (CDA 122) is prone to both embrittlement and IGSCC. Copper-phosphorus alloys are most susceptible to IGSCC at approximately 0.2% phosphorus content.

TGSCC of aluminum bronze (similar to CDA 613) is known to occur in moist ammonia-containing environments. Aluminum bronze having aluminum contents intermediate to those of CDA 102 and CDA 613 (approximately 1% aluminum) is more susceptible to SCC than aluminum bronze of other compositions. IGSCC of aluminum bronze is known to occur in steam environments, probably because of the formation of Al_2O_3 at grain boundaries, but it can be effectively inhibited with small additions of tin. The tin impedes the oxidation of intergranular aluminum. Aluminum bronze (CDA 613) is very

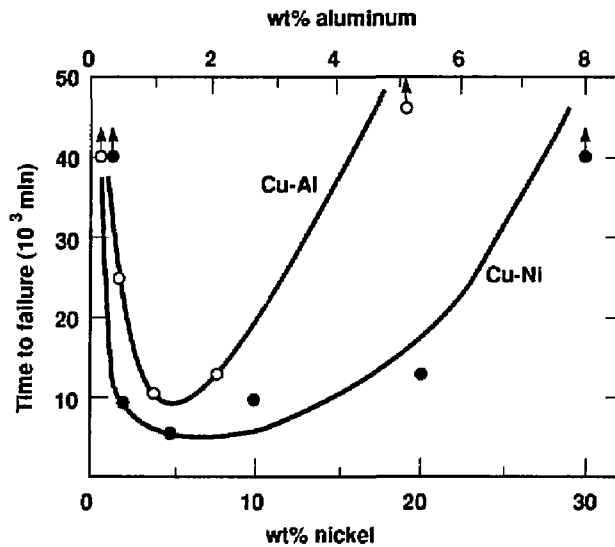


Figure 17. SCC of copper-based alloys in moist ammonia vapor during constant-load testing [19]. The compositions of CDA 613 and CDA 715 have been optimized to decrease susceptibility to SCC.

susceptible to dealuminification, which could conceivably induce failure by a film-induced cleavage SCC mechanism. Chloride and sulfate anions may promote dealloying of materials such as CDA 613; however, these ionic species do not appear to promote SCC directly. This behavior is in contrast to that of the austenitic candidate alloys.

IGSCC of cupronickel alloys such as Cu-30Ni (CDA 715) may occur in steam environments, but these alloys are much less susceptible to IGSCC than aluminum bronzes (without tin inhibitor). TGSCC of cupronickel alloys may occur in moist, ammonia-containing environments, but these alloys are much less susceptible to TGSCC than aluminum bronzes (CDA 613). The passive films on cupronickel alloys probably consist of $\text{Ni}(\text{OH})_2$. Auger spectroscopy has shown that these films primarily contain nickel and oxygen (note that hydrogen cannot be detected by Auger spectroscopy). Similar films would be expected at the crack tip. Though some dealloying may occur in cupronickel alloys (CDA 715), the rates of dealloying are several orders of magnitude smaller than the rates of dealloying observed in aluminum bronzes (CDA 613). Therefore, TGSCC by the postulated mechanism of film-induced cleavage in dealloyed layers would be expected to be a more significant problem in CDA 613 than in

CDA 715, though relatively few data exist supporting such TGSCC failures in either material.

It is generally accepted that all copper alloys are resistant to marine biofouling, as evidenced by the vast number of applications of these materials in marine environments. However, evidence does exist which indicates that these materials are susceptible to some types of MIC.

The only data found in the literature that allow a direct comparison of the SCC susceptibilities of the copper-based candidate alloys were published by Pugh et al. [20]. These data are not specifically for CDA 613 and CDA 715 but for alloys having close compositions, i.e., Cu-25Ni and Cu-6Al. Furthermore, these data are only applicable to moist ammonia-containing environments. The data are summarized in Table 3.

The following tentative ranking can be inferred for susceptibility to SCC in *tarnishing* ammoniacal solutions:

CDA 715 = CDA 102 (best) > CDA 613 (worst)

Correlations between corrosion potential and susceptibility to SCC have been established for neutral Mattsson solution that also suggest that CDA 715 is superior to CDA 613. This ranking is in contrast to that for *nontarnishing* ammoniacal solutions:

Table 3. Time-to-failure of copper-based alloys in ammoniacal solutions [20].

Solution	Cu	Cu-6Al	Cu-25Ni
Tarnishing	>100,000	2,600	>100,000
Nontarnishing	4,600	16,700	11,900

Note: Time in seconds; specimens were stressed at 10% of proof stress.

CDA 715 = CDA 613 (best) > CDA 102 (worst)

It may be possible for SCC to originate in dealloyed layers and propagate by a film-induced cleavage mechanism. In such cases, susceptibility to SCC should be related to dealloying kinetics. On the basis of data from the literature on dealloying in chloride and sulfate environments, the following ranking can be deduced:

CDA 715 (better) > CDA 613 (worse)

Obviously, CDA 102 would not fail by this type of mechanism.

It can be argued, on the basis of the limited data available, that CDA 715 has the best overall resistance to SCC. There is a clear need for additional experimental data, however. Sufficient quantitative data do not exist for the unambiguous ranking of these materials under other conditions that may be more relevant to the repository environment, such as exposure to steam and nitrite.

5. Localized Corrosion of Copper-Based Alloys

In Volume 5 [21], Farmer et al. have conducted a critical survey of the available literature (123 articles) on pitting and crevice corrosion of the copper-based alloys. Pitting and crevice corrosion are two of the most common forms of LC of copper-based alloys. Pitting usually occurs in water that contains low concentrations of hydrogen carbonate (bicarbonate) and chloride anions, such as water from Well J-13 at the Nevada Test Site. Consequently, this mode of degradation might occur in the repository environment.

The copper-based candidate materials are sensitive to several species that may exist in the repository environment. Ions known to play an important role in pitting include S^{2-} , HS^- , HCO_3^- , SO_4^{2-} , Cl^- , Fe^{3+} , and H^+ . Dissolved O_2 is also very important. In addition to the reduction of oxygen, the reduction of ferric ion to ferrous ion can serve to depolarize anodic reactions involved in pitting and SCC. The type of pitting observed on copper depends on the $[HCO_3^-]:[SO_4^{2-}]$ ratio, so measurements of sulfate concentration are important. At the repository, we expect roughly 120 ppm carbonate and 20 ppm sulfate, so the ratio will be much greater than 1.

Two types of pitting are seen in fresh water. Type 1 pitting is usually associated with certain

hard or moderately hard well waters. This type of pitting is believed to be due to a continuous carbon film formed on the inner surface of tubes during bright annealing. It is more likely to occur in cold water than hot water and may cause perforation of tubes after only 1 yr of exposure. It is characterized by the formation of fairly large, well-defined pits, usually containing soft crystalline cuprous oxide, and often cuprous chloride, beneath hard green mounds of calcium carbonate and basic copper carbonate. The surface between the pits is often covered with a shiny, dark red, water-formed cuprous oxide layer beneath a greenish deposit of hardness salts stained with traces of copper corrosion products. On the basis of the expected carbonate to sulfate ratio, we would expect this type of pitting, but only when the temperature decreases to well below 60°C. This decrease might occur only many hundreds of years after emplacement in the repository.

Type 2 pitting occurs on hard-drawn tubes in hot tap water of low pH (<7.4) and a low $[HCO_3^-]:[SO_4^{2-}]$ ratio (<1). This type of pitting is practically unknown if the water temperature is below 60°C (140°F). It is characterized by deep pits, of small cross section, containing very hard crystalline cuprous oxide and capped by small black or greenish-black mounds of cuprous oxide

and basic copper sulfate. On the basis of the carbonate to sulfate ratio, we would not expect this type of pitting to occur.

Crevice corrosion is a form of localized attack that occurs at shielded areas on metal surfaces exposed to certain environments. The sites for this type of corrosion are often unavoidable because of the structural or functional design and may even arise during the exposure period. Examples of sites include spot-welded lap joints, threaded or riveted connections, gasket fittings, porous welds, valve seats, coiled or stacked sheet metal, marine or debris deposits, and the water line.

The penetration of corrosive solutions into these relatively inaccessible areas, with widths that are typically a few thousandths of an inch, can result in various types of failures: the metal surface may become stained or perforated by the corrosive, the mechanical strength may be reduced below tolerance limits so that fracture occurs from the applied load or from the wedging action of the corrosion products, operating components may seize, or protective coatings may be disbonded from the metal surface.

Much of the theoretical interpretation of crevice corrosion has been associated with a concentration cell, wherein aggressive solute species in water become highly concentrated at the crack tip. This concept is based on the fact that the initiation and propagation of localized attack on metal surfaces are influenced by nonuniformity of the corrosive solution. For example, variations in the concentration of ions or dissolved gases may

arise along the same metal surface because of the existence of crevices that are relatively inaccessible to fresh electrolyte compared with freely exposed surfaces. Such environmental conditions cause potential differences and influence electrode kinetics to the extent that an electrochemical cell is established, wherein an oxidation process (that is, corrosion) occurs at the anodic sites, and some reduction process (for example, oxygen reduction) occurs at the cathodic sites.

A ranking by Tuthill and Schillmoller [22] of metals and alloys in terms of resistance to crevice corrosion in quiet seawater is given in Table 4. Though few quantitative LC data were found, qualitative information was used to establish the following tentative ranking:

Pitting corrosion
 CDA 102 (best) = CDA 715 < CDA 613 (worst)
 Local dealloying
 CDA 102 (best) > CDA 715 > CDA 613 (worst)
 Crevice corrosion
 CDA 715 (best) = CDA 613 > CDA 102 (worst)
 Biofouling
 CDA 102 (best) > CDA 715 = CDA 613 (worst)

The susceptibility of CDA 102 to crevice corrosion makes it a less suitable choice even though it performs well in the categories of pitting corrosion, local dealloying, and biofouling. The cupronickel alloy, CDA 715, probably has the best overall resistance to such localized attack.

Table 4. Relative crevice-corrosion resistance of metals and alloys in quiet seawater [22].

Metal or alloy	Resistance
Cu-30Ni-0.5Fe (CDA 715)	Best—very little attack
Aluminum bronzes (CDA 613)	Best—very little attack
Alloy 825	Less—significant attack
Copper (CDA 102)	Less—significant attack
Type 316 stainless steel	Pit initiation at crevice
Type 304 stainless steel	Pit initiation at crevice

6. Effects of Hydrogen in Austenitic and Copper-Based Alloys

In Volume 6 [23], Gdowski and Bullen have surveyed the literature (86 articles) on the effect of hydrogen in the candidate materials. Hydrogen can be generated by electrochemical corrosion reactions.

The following points are made in regard to the effects of hydrogen in the candidate alloys. First, the absorption of hydrogen in both the copper-based and austenitic alloys is not spontaneous. Deformation must be applied for the reaction to occur. Second, the solubility and diffusivity of hydrogen are such that the reaction does not occur at even a slightly elevated temperature. Third, hydrogen diffusing along grain boundaries can potentially react with constituent phases in copper-based or austenitic alloys to form gas-filled voids. The presence of these voids degrades mechanical properties. For example, absorbed and diffusing hydrogen reacts with cuprous oxide in copper to form water vapor (*Wasserstoffkrankheit*). Deoxidizers in copper, such as phosphorus, can reduce susceptibility to hydrogen. An increased concentration of nickel enhances the solubility of hydrogen in cupronickel alloys, thereby decreasing the susceptibility of Cu-30Ni (similar to CDA 715) to hydrogen embrittlement.

The copper-based alloys are ranked in order of their resistance to this mode of degradation as follows:

CDA 715 (best) > CDA 613 > CDA 102 (worst)

In contrast to its effects in copper-based alloys, absorbed and diffusing hydrogen reacts with carbon in austenitic stainless steels to form methane. Hydrogen can also precipitate as hydrogen bubbles in cold-worked materials with a high dislocation density. These processes can be prevented by avoiding fabrication methods and alloys that result in martensite formation.

The austenitic alloys are ranked in order of their resistance to this mode of degradation as follows:

Type 316L (best) > Alloy 825 > Type 304L (worst)

To put this discussion in proper perspective, we do not believe that this mode of degradation will limit container lifetimes given the alloys, container temperatures, environment, and fabrication methods we intend to use.

7. Weldability of Austenitic Alloys

In Volume 7 [24], Strum et al. discuss problems specifically associated with welding the austenitic candidates, which are Types 304L and 316L stainless steels and Alloy 825. The problems discussed include weld-induced sensitization, hot cracking, σ -phase formation, and corrosion of weld filler metal.

Sensitization is due to the formation of $Cr_{23}C_6$ carbides at grain boundaries and the depletion of chromium near precipitates. Such carbide formation can be prevalent in the heat-affected zone (HAZ) of welds. Control of carbon content in Type 304L or 316L to less than 0.035% is effective in minimizing the sensitization. In Alloy 825, the problem is effectively eliminated by the addition of titanium, which promotes formation of titanium carbides, thus minimizing the loss of chromium. The reader is referred to the discussions of sensitization in Sections 1 and 3 above as well as in Vols. 1 and 3 of the survey [3, 10].

Hot cracking in the HAZ and in the weld zone (WZ) is also reviewed in Vol. 7. The ferrite phase that is formed in the HAZ and WZ provides a positive countermeasure against grain-boundary cracking due to segregation of impurities. This phase is believed to act as an impurity sink. Furthermore, the presence of manganese in the ferrite prevents the formation of low-melting-point eutectics at grain boundaries by the formation of intermetallic compounds. Note that the weld metal compositions in both Types 304L and 316L stainless steels are adjusted to assure a content of approximately 5% ferrite. This is not possible in Alloy 825 since it is purely austenitic. The most generally accepted method of preventing hot cracking in Alloy 825 is to substitute a weld filler material of a different composition, such as Alloy 625.

Alloy 625 is more noble than Alloy 825, so any galvanic corrosion will be in the Alloy 825 that is in close proximity to the weld metal. It is

not possible to treat the entire Alloy 825 surface as an anode because of the primary current distribution in the vicinity of the weld. This phenomenon presents a potential problem because the HAZ could therefore suffer LC. Since Alloy 625 has more molybdenum than Alloy 825, it has a more stable passive film. Consequently, Alloy 625 is more resistant to crevice corrosion and pitting than Alloy 825 [14]. This resistance is reflected in the pitting potentials shown in Fig. 18. Alloy 625 has exhibited no SCC under conditions known to be detrimental to alloys such as Types 304L and 316L.

Phase stability of the weld metal is also an important consideration. Thermodynamically stable single-phase materials are desirable since phase changes can result in degradation of mechanical properties. The hard, brittle, nonmagnetic σ phase that forms at elevated temperatures and under certain conditions in alloys such as Types 304L and 316L remains stable at room temperature. The σ phase can result in the degradation of some mechanical properties, namely, fracture toughness or impact resistance. Data have been found that suggest that σ phase can form during the welding of alloys such as Types 304L

and 316L. However, considering the high nickel content of Alloy 825, it is believed that the potential for the formation of a brittle intermetallic phase in Alloy 825 is highly unlikely. Alloy 625 is a nickel-chromium-molybdenum alloy with a small amount of iron, manganese, silicon, aluminum, and columbium or tantalum. Investigation of the stability of Alloy 625 following extended heat treatments in the temperature range of 593 to 927°C indicates no intermetallic phase formation [26]. The composition of Alloy 625, determined using the calculated equivalent-chromium concentration, is also shown in Fig. 1. From the calculated equivalent-chromium concentration, it appears that this alloy could exist in either the austenite (γ) region or in a two-phase region where austenite and ferrite (α) would coexist. Welding with Alloy 625 filler could lead to such a duplex microstructure.

On the basis of resistance to welding-induced degradation, the austenitic materials are ranked as follows:

Alloy 825 (best) > Type 316L stainless steel > Type 304L stainless steel (worst)

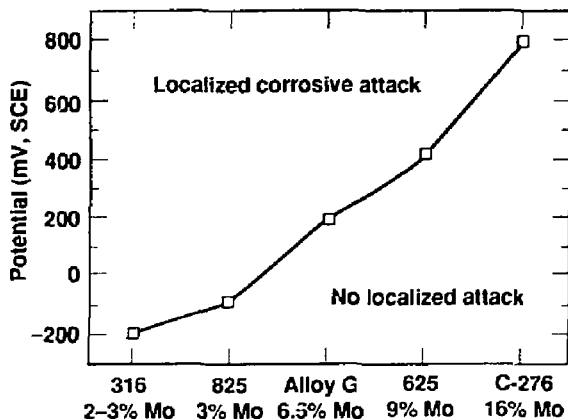


Figure 18. Effect of molybdenum alloy additions on the LC potential for a constant applied potential, 24-hr exposure in 3.8% FeCl_3 at 70°C [25].

8. Weldability of Copper-Based Alloys

In Volume 8 [27], Bullen et al. review the technical literature on the welding of the copper-based candidate materials CDA 102, CDA 613, and CDA 715. While their review identifies a number of potential problems, the problems do not appear to preclude the application of these materials in the fabrication of containers for the storage of high-level radioactive waste.

CDA 102 is difficult to weld since very high heat input is required to compensate for its high thermal conductivity. The significant preheat requirements of conventional welding techniques lead to excessive oxidation of the material. The use of an inert shield gas is required to prevent oxide formation and the associated reduction of cuprous oxide by hydrogen, which results in the formation of water vapor and porosity. The use of a deoxidized filler material, or, possibly, the consideration of a deoxidized grade of high-purity copper such as CDA 122, may be necessary if high-purity copper is selected as the metal barrier material.

CDA 613 appears to have low ductility in the temperature range of 400 to 600°C. Brittle failure over this temperature range is thought to be a result of the segregation and precipitation of impurity elements during the welding process. This segregation and embrittlement phenomenon is exacerbated by the repeated thermal cycling produced in multipass welds. Welding of aluminum bronze alloys also requires the use of inert gas shielding to prevent the formation of an aluminum oxide scale, which is insoluble in most flux materials. These oxide particles can become entrapped in the melt and result in weld embrittlement. Aluminum bronze welds have also been shown to dealloy in an acid corrosion environment over time. This dealloying could lead to significant corrosion problems if it occurred in a repository environment.

CDA 715 can be subject to problems associated with dealloying. Fusion-line cracking is seen in cupronickel welds as a result of segregation of major alloying elements and impurities. Partial melting, associated with localized solute segregation, is a probable cause of hot cracking in this material. Trace elements, such as lead, sulfur, bismuth, selenium, phosphorus, and tellurium can be tolerated with the use of nickel filler wire under proper welding conditions. When proper inert gas shielding is not employed, cupronickels are also identified as being prone to atmospheric

contamination, which causes porosity in weldments. Because of a relatively wide solid-solution freezing range, cupronickels can also exhibit coring, which results in subsolidus tearing, possibly leading to cracking.

Alternative welding methods that limit heat input and yield a relatively small HAZ are also reviewed in Vol. 8. Electron-beam welding requires low total heat input and is therefore identified as a possible method of limiting solute segregation and partial melting arising from impurity eutectic formation. Electron-beam welding is completed in a vacuum, thus eliminating the need for a shielding gas to prevent porosity from hydrogen or water-vapor entrapment. In addition, electron-beam welding is an autogenous process requiring no filler material. Inertial or friction welding is also identified as a potential joining alternative. This process is relatively insensitive to part configuration and welding parameters. There is no fusion on the macroscopic scale and therefore no traditional fusion zone. Solute segregation is precluded since there is no melt zone. However, very large equipment is required and considerable study must be conducted before this approach can be qualified for use in a hot cell.

In summary, CDA 715 is judged to be superior since it undergoes less catastrophic degradation and failure with respect to oxide formation and loss of ductility. The microsegregation of alloying elements and impurities and the partial melting documented in Vol. 8 may be overcome with the application of alternative welding techniques, such as electron-beam and inertial or friction welding. CDA 102, though prone to the effects of "hydrogen sickness" or formation of porosity due to the reduction of entrapped cuprous oxide particles, could be employed as a container material if a suitable filler material with sufficient deoxidizing capabilities were used. The utilization of an alternate high-purity copper alloy, such as CDA 122, may preclude this requirement. CDA 613 appears to have the most significant problems when welding is considered. The formation of an aluminum oxide scale and the inclusion of oxide particles because of inadequate inert gas shielding could significantly affect the integrity of welds. The possible loss of ductility in the temperature range of 400 to 600°C, and the resulting possibility of hot cracking, suggest that difficulties could arise in the fabrication of high-level nuclear-waste containers if CDA 613 is used.

Considering the effects of welding alone, the following ranking of the copper-based candidate

materials is proposed:
CDA 715 (best) > CDA 102 > CDA 613 (worst)

Conclusions and Future Work

The candidates are tentatively ranked on the basis of this survey. Alloy 825 and CDA 715 are believed to have the best overall performance. Alloy 825 is a metallurgically stable austenite, whereas Types 304L and 316L stainless steels are metastable and take on duplex austenite-ferrite structures under thermal-equilibrium conditions. CDA 715 (70-30 copper-nickel) will probably exist as a simple solid solution of nickel in copper, although thermodynamic calculations have been used to predict a two-phase region. CDA 715 does not suffer from the internal oxidation problems encountered with CDA 102 (oxygen-free copper). The mechanical strength of CDA 613 (aluminum bronze) depends on iron precipitates, which may grow in size over extended periods of time.

Although pitting and crevice corrosion may be encountered with all of the candidates, Alloy 825 and CDA 715 seem the least susceptible to these forms of attack. Under environmental conditions known to induce SCC of Types 304L and 316L stainless steels, Alloy 825 appears resistant to this mode of degradation. Similarly, CDA 715 is the only copper-based alloy that appears to have good resistance to SCC over a broad range of conditions known to cause failure in CDA 102 and CDA 613.

Types 304L and 316L stainless steels are probably easier to weld than Alloy 825. CDA 715 appears to be the best copper-based candidate in terms of weldability.

It is important to note that the rankings in this report are based on information in the general technical literature. While the literature includes a broad range of environmental conditions, the rankings are not based on experimental work specific to Yucca Mountain. We have tried to use available information that fits within the brackets formed by the physical, chemical, metallurgical, and mechanical conditions that are expected to occur during the containment period at Yucca Mountain. There are important gaps between the conditions published in the literature and the conditions expected at Yucca Mountain. For instance, the combined effect of a moderately high temperature, high radiation field, and ambient oxidizing conditions on the metal oxidation and corrosion rates is not known. The analysis of

information from the surveys indicates that this lack of correspondence is particularly important in the copper-based materials and less important in the austenitic materials. Much of the data on LC of the copper-based materials appears to be more empirically oriented than otherwise comparable data on the austenitic materials. In the austenitic materials, a higher degree of correlation exists between experimental and field data on the one hand and mechanistic models for predicting behavior on the other.

Although a tentative ranking of the candidate alloys is possible between members of the same alloy family (because alloy composition is often used as an experimental parameter), a comparison between candidates in the two alloy families has not yet been made. This comparison is needed as part of the selection process because the plan of the Metal Barrier Selection and Testing Task is to nominate one of the six candidates for future design evaluation. In the next several months, we plan to (1) perform electrochemical determinations of critical LC potentials and incubation times for all of the candidate materials, and (2) generate data on detecting the initiation and propagation of stress corrosion in all of the candidate materials, including weldments. The welding processes that we will study represent those under consideration for making the closure weld of the containers. Test environments will be relevant to the repository, although specific aspects of the chemical environment will be modified to accelerate degradation so that data can be obtained in a reasonable amount of time. Specific heat treatments will also be given to the specimens to accelerate aging or transformation phenomena in order to better simulate actual as-emplaced conditions. Completion of this activity will allow a direct comparison of the behavior of the candidates.

Some experimental work conducted under conditions relevant to the environment, temperature, and radiation conditions of the repository has been completed and published [8]. Additional test data will be published in the coming months.

Factors other than those related to material performance will enter into the selection process. These factors include practical concerns about

fabricating, closing, and inspecting the disposal containers. The constraints of doing many of these operations remotely in a hot cell must be incorporated into process considerations. There are, obviously, close interactions between the corrosion behavior of materials and the processes used for fabricating and joining them; some of these interactions are discussed in the volumes on the effects of welding on degradation (Vols. 7 and 8).

Other parts of the NNWSI Project are currently evaluating the fabrication, closure, and inspection processes. Compatibility with the waste form and with other waste-package components is also being evaluated elsewhere in the Project. Material-related costs in producing, handling, and emplacing the container are also a factor in selection. Again, other parts of the Project are aiding in the evaluation of these concerns. Last, a technological data base showing where the candidate materials have been successfully used in engineering applications is considered important in the selection process. We are drawing on the ex-

periences of many people in the metallurgical engineering profession to help construct this data base.

All of these different factors will be elements of the criteria to be used in selection of the container material. The Metal Barrier Selection and Testing Task is in the process of developing a formal selection process. First, the selection criteria will be established, including the weighting factors and how individual criteria will be scaled. Then, each candidate material will be evaluated against the criteria and scored. It is proposed that there be a minimum passing score for each criterion. The material with the top total score, provided it satisfies the minimum requirement for each criterion, will be the one recommended to the NNWSI Project for continued evaluation during the period of future design work and the period of data acquisition for license application. The series of surveys of degradation modes that this overview introduces is a key part of the information that will be used in container-material selection.

Acknowledgments

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Appendix

Methodology of the Literature Search

An extensive search of the technical literature was performed in order to obtain the necessary information for a thorough discussion of the degradation modes of the candidate container materials under repository-relevant conditions. The primary source of information was Lawrence Livermore National Laboratory's (LLNL's) Technical Information Department Library. The methods employed in the review of the technical literature included:

1. Computer searches of applicable data bases to identify relevant articles and reviews. (Tables A1 and A2 list the computer searches performed.)
2. Acquisition and study of review articles that provided overview information on a topic.
3. Obtaining references, identified in the review articles, that might be applicable to repository-relevant materials and conditions.
4. Review of conference proceedings, such as those of the National Association of Corrosion Engineers (NACE).
5. Review of books, such as the American Society for Testing and Materials' Special Technical Publications and books on specific topics.
6. Review of the current literature for data not yet cataloged in data bases. This literature included the following journals: *Corrosion*, *Materials Performance*, *Corrosion Reviews*, *Corrosion Science*, *The British Corrosion Journal*, *Journal of Materials Science*, and *Journal of Nuclear Materials*.
7. Obtaining references identified in the articles reviewed. Data bases do not generally catalog articles before the 1960s; articles published before then were obtained from citations in more recent articles.
8. Manual search of selected journals for articles. The search involved a review of the subject indexes of the journals.

Articles and books not obtainable directly from the LLNL Technical Information Department Library were obtained through the LLNL library acquisition system.

It should be noted that completion of a technical literature search is an iterative process. Each article obtained may contain relevant references. Hence, it is necessary to trace references from numerous sources in hopes of identifying most of the available data on a given subject.

Table A1. On-line data bases used in literature searches of austenitic materials.

Data base, search date	Search identifiers	Number of items identified
Stress corrosion cracking		
Metadex (1966–present), 23 Sept. 1987	Stress (w) Corrosion (w) Cracking and Stainless (w) Steel and 304#	a 2783 Sum 489 abstracts
Metadex (1966–present), 23 Sept. 1987	Stress (w) Corrosion (w) Cracking and Stainless (w) Steel and 316#	a 2055 Sum 117 abstracts
Metadex (1966–present), 22 Sept. 1987	Stress (w) Corrosion (w) Cracking or Crevice (w) Corrosion or Corrosion (w) (Mechanism or Rate or Potential) and Alloy (w) 825	a a a 12 Sum 5 abstracts
Modeling—Stress Corrosion Cracking		
Metadex (1966–1987/July) File 32, 1 July 1987	(Stress () Corrosion () Cracking and (Simulation? ? or Modelling or Modeling or Model? ?))//TI,DE	a a a Sum 116 abstracts
Dialog Aerospace (62–87) File 108, 1 July 1987	(Stress () Corrosion () Cracking and (Simulation? ? or Modelling or Modeling or Model? ?))//TI,DE	a a a Sum 97 abstracts
NTIS (64–87) File 6, 1 July 1987	(Stress () Corrosion () Cracking and (Simulation? ? or Modelling or Modeling or Model? ?))//TI,DE	a a a Sum 28 abstracts
World Aluminum Abstracts (1968–1987/June) File 33, 1 July 1987	(Stress () Corrosion () Cracking and (Simulation? ? or Modelling or Modeling or Model? ?))//TI,DE	a a a Sum 21 abstracts

^a Number was not provided by the search.

Table A1. (Continued.)

Data base, search date	Search identifiers	Number of items identified
Crevice Corrosion		
Metadex (1966–present), 23 Sept. 1987	Crevice (w) Corrosion and Stainless (w) Steel and 304#	^a 2783 Sum 55 abstracts
Metadex (1966–present), 23 Sept. 1987	Crevice (w) Corrosion and Stainless (w) Steel and 316#	^a 2055 Sum 64 abstracts
Metadex, 22 Sept. 1987	Stress (w) Corrosion (w) Cracking or Crevice (w) Corrosion or Corrosion (w) (Mechanism or Rate or Potential) and Alloy (w) 825	^a ^a ^a 12 Sum 5 abstracts
Pitting		
Metadex (1966–present), 23 Sept. 1987	Pitting and Stainless (w) Steel and 304#	^a 2783 Sum 181 abstracts
Metadex (1966–present), 23 Sept. 1987	Stress (w) Corrosion (w) Cracking and Stainless (w) Steel and 316#	^a 2055 Sum 118 abstracts
Corrosion Mechanisms, Rates, Potential—Austenitics		
Metadex (1966–present), 23 Sept. 1987	Corrosion (w) (Mechanism or Rate or Potential) and Stainless (w) Steel and 304#	^a 2783 Sum 167 abstracts
Metadex (1966–present), 23 Sept. 1987	Corrosion (w) (Mechanism or Rate or Potential) and Stainless (w) Steel and 316#	^a 2055 Sum 91 abstracts

^a Number was not provided by the search.

Table A1. (Continued.)

Data base, search date	Search identifiers	Number of items identified
Metadex (1966–present), 22 Sept. 1987	Stress (w) Corrosion (w) Cracking or Crevice (w) Corrosion or Corrosion (w) (Mechanism or Rate or Potential) and Alloy (w) 82	^a ^a ^a 512 Sum 5 abstracts
Oxidation and Corrosion—Alloy 825		
Metadex (1966–present), 23 June 1987	Incoloy (w) 825 or Alloy (w) 825 or Ni (w) O (w) Nel or Incoloy (w) 825 or Alloy (w) 825 and STE Corrosion or Oxidation	43 12 0 54 72,073 Sum 49 abstracts
Energy (87–76), 23 June 1987	Incoloy (w) 825 or Alloy (w) 825 or Ni (w) O (w) Nel and STE Corrosion or Oxidation	^a ^a ^a ^a Sum 50 titles
Metadex (1966–present), 10 Feb. 1988	"Corrosion Incoloy 825"	Sum 51 abstracts
Chemical Abstracts, 10 Feb. 1988	"Corrosion Incoloy 825"	Sum 22 abstracts
Chemical Abstracts, 10 Feb. 1988	"Corrosion Incoloy 825"	Sum 72 abstracts
INSPEC, 10 Feb. 1988	"Corrosion Incoloy 825"	Sum 12 abstracts
NTIS, 10 Feb. 1988	"Corrosion Incoloy 825"	Sum 26 abstracts
Compendex (88–72), 10 Feb. 1988	"Corrosion Incoloy 825"	Sum 35 abstracts

^a Number was not provided by the search.

Table A1. (Continued.)

Data base, search date	Search identifiers	Number of items identified
Phase Stability and Hydrogen Embrittlement		
Metadex (1966–present), 5 Nov. 1986	Austenitic (L) Stainless (L) Steel and Phase (w) Stability or Hydrogen (w) Embrittlement	3268 6236
		Sum 124 titles
Metadex (1966–present)	Incoloy (2w) 825 or Incoloy—825 and Phase (w) Stability	^a 206 1937
		Sum 3 abstracts
Compendex	Phase (l) Stability or Hydrogen (2N) Effect or Hydrogen (l) Embrittlement and Incoloy	326 2460 316
		Sum 8 abstracts
EI Meeting	Incoloy or Incoloy (2w) 825 and Phase Stability or Hydrogen (2N) Effect or Hydrogen (l) Embrittlement	163 144 885
		Sum 9 abstracts
DOE Energy Files 103, 104	Incoloy or Incoloy (2w) 825 and Phase Stability, or Hydrogen (2N) Effect or Hydrogen (l) Embrittlement	^a ^a ^a
		Sum 10 abstracts
NTIS 6	Incoloy or Incoloy (2w) 825 and Phase Stability or Hydrogen (2N) Effect or Hydrogen (l) Embrittlement	^a ^a ^a
		Sum 6 titles

^a Number was not provided by the search.

Table A1. (Continued.)

Data base, search date	Search identifiers	Number of items identified
Aerospace 108	Incoloy or Incoloy (2w) 825 and Phase Stability or Hydrogen (2N) Effect or Hydrogen () Embrittlement	a a a
		Sum 5 abstracts
DOE Energy (74-82), 26 Jan. 1987	"Stainless Steel Sensitization"	Sum 28 abstracts
Metadex (1966-present), 27 Jan. 1987	"Stainless Steel Sensitization"	Sum 52 abstracts
Welding		
Metadex, 24 Feb. 1988	304/ALI and Welding/CT and Corrosion/CT or Crack?/CT or Sensitization or Stress/CT	5669 566 a a a
		Sum 54 abstracts

^a Number was not provided by the search.

Table A2. On-line data bases used in literature searches of copper and copper alloys.

Data base, search date	Search identifiers	Number of items identified
Crevice Corrosion or Pitting or Stress Corrosion Cracking		
Metadex (1966-present), 16 Feb. 1988	(Crevice (w) Corrosion or Pitting or Stress (w) Corrosion (w) Cracking and Cu-30Ni or Cu-7Al or 613/AlI or 715/AlI or 102/AlI or (Copper or Cu) (a) OFHC	13,198
		193
		35
		29
		9
		156
	Sum 33 abstracts	
INSPEC, 16 Feb. 1988	Crevice (w) Corrosion or Pitting or Stress (w) Corrosion (w) Cracking and Cu*Ni/ET or Al*Cu/ET or (Copper or Cu) (a) OFHC	4695
		1554
		2436
		205
		Sum 56 titles
Energy, 16 Feb. 1988	Crevice (w) Corrosion or Pitting or Stress (w) Corrosion (w) Cracking and or (Copper OR Cu) (a) OFHC	a
		a
		a
		a
		a
		Sum 2 abstracts
Energy, 16 Feb. 1988	Crevice (w) Corrosion or Pitting or Stress (w) Corrosion (w) Cracking and Cu*Ni/ET or Al*Cu/ET	a
		a
		a
		a
		a
		Sum 11 abstracts
Chemical Abstracts, 16 Feb. 1988	Crevice (w) Corrosion or Pitting or Stress (w) Corrosion (w) Cracking and CDA (w) 613 or CDA (w) 715	a
		a
		a
		a
		a
		Sum 6 abstracts

^a Number was not provided by the search.

Table A2. (Continued.)

Data base, search date	Search identifiers	Number of items identified
Metadex (1966–present), 1 April 1987	Corrosion	37,777
	and	
	Copper (a) Nickel or Cu (2a) Ni	6745
	and	
	Alloy	102
	not Cr	67
	not Steel	58
	not (Zinc or Zn)	50
		Sum 50 abstracts
Pitting and Crevice Corrosion		
Chemical Abstracts, 6 May 1988	CDA and 715	a
	and	
	Pitting or Crevice (w) Corrosion	a
		Sum 4 abstracts
Chemical Abstracts, 6 May 1988	Copper and OFHC	a
	and	
	Pitting or Crevice (w) Corrosion	a
		Sum 3 abstracts
Metadex (1966–present), 6 May 1988	613/ALI	6
	and	
	Pitting or Crevice (w) Corrosion	5168
		Sum 1 abstract
INSPEC, 6 May 1988	Al*Cu/ET or Cu-Al/ET	2471
	and	
	Pitting or Crevice (w) Corrosion	a
		Sum 17 abstracts
INSPEC, 6 May 1988	Cu*Ni/ET	1572
	and	
	Pitting or Crevice (w) Corrosion	a
		Sum 6 abstracts
INSPEC, 6 May 1988	Cu Et/CHI or OFHC	2475
	and	
	Pitting or Crevice (w) Corrosion	a
		Sum 8 abstracts

^a Number was not provided by the search.

Table A2. (Continued.)

Data base, search date	Search identifiers	Number of items identified	
Stress Corrosion Cracking, Crevice Corrosion, and Corrosion Mechanisms			
Metadex (1966-present), 22 Sept. 1987	CDA (w) 102	1	
	or 102/ALI	9	
	or CDA 102/ALI	1	
	or CDA 613/ALI	0	
	or CDA 715/ALI	2	
	or Cu-30Ni/ALI	120	
	or Cu-7Al/ALI	34	
	or OFHC and Copper	158	
	and		
	Stress (w) Corrosion (w) Cracking or Crevice (w) Corrosion or Corrosion (w) Mechanism	7518 810 a	
	Sum 29 abstracts		
Corrosion			
Metadex (1966-present), 14 Sept. 1987	Corrosion and High (a) Purity (a) Copper	60,252 82 Sum 1 abstract	
	Metadex (1966-present), 14 Sept. 1987	Corrosion/CT and Cupronickel/CT	39,137 182 Sum 90 titles
		Metadex (1966-present), 14 Sept. 1987	Corrosion/CT and Aluminum Bronzes/CT
Metadex (84-87), 31 March 1987			Corrosion and Copper (w) Aluminum or Copper (w) Aluminium and Alloy
	Metadex (84-87), 31 March 1987		Corrosion and CDA 715 or CDA 613 or CDA 102

^a Number was not provided by the search.

Table A2. (Continued.)

Data base, search date	Search identifiers	Number of items identified
Compendex, 22 Sept. 1987	OFHC (w) Copper or CDA (w) (102 or 715 or 613) or Cu (w) 30Ni or Cu (w) 7Al and Corrosion	166 3 9 1 a Sum 6 abstracts
Chemical Abstracts, 22 Sept. 1987	OFHC (w) Copper or CDA (w) (102 or 715 or 613) or Cu (w) 30Ni or Cu (w) 7Al and Corrosion	a a a a a Sum 3 abstracts
Hydrogen Effects and Embrittlement		
DOE Energy (1974–1982, 1983–1986) Files 103 and 104, 27 Jan. 1987	"Hydrogen Effects in Cu"	Sum 16 abstracts
Nuclear Science Abstracts (NSA) (1948–1976) File 109, 27 Jan. 1987	"Hydrogen Effects in Cu"	Sum 6 abstracts
NTIS (64–87), 27 Jan. 1987	"Hydrogen Effects in Cu"	Sum 7 abstracts
INSPEC (1969–1976, 1977– present) Files 12, 13, 27 Jan. 1987	"Hydrogen Effects in Cu"	Sum 21 abstracts
Aerospace (62–87), 27 Jan. 1987	"Hydrogen Effects in Cu"	Sum 1 abstract
Metadex (1966–present)	"Hydrogen Embrittlement of Cu, Cu Alloys"	Sum 55 abstracts
Chemical Abstracts (1967– 1986) Vol. 105	"Hydrogen Embrittlement of Cu, Cu Alloys"	Sum 27 titles

^a Number was not provided by the search.

Table A2. (Continued.)

Data base, search date	Search identifiers	Number of items identified
Phase and Precipitate Stability		
Metadex (1966–present), 9 Dec. 1986	“Phase and Precipitate Stability in Cu Alloys”	Sum 40 abstracts
Chemical Abstracts (1967– 1986) File 105, 9 Dec. 1986	“Phase and Precipitate Stability in Cu Alloys”	Sum 110 titles

^a Number was not provided by the search.

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