OVERVIEW OF C-22 ALLOY (UNS N06022) FOR USE IN HIGH LEVEL WASTE PACKAGE CANISTERS

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ABSTRACT

C-22 alloy has been selected for use in the waste package canisters at the Yucca Mountain site. This study shows that thermal aging of this material, as might occur from heat produced by the radioactive waste, will not degrade the performance of the C-22 alloy, even for time periods up to 10,000 years. Microstructural, mechanical, and corrosion measurements made on C-22 alloy specimens aged at various temperatures up to 40,000 hours have been used to demonstrate its thermal stability. In addition, extrapolation of the higher temperature data to longer times (10,000 year) and lower temperatures (below 350°C) show that the C-22 alloy will maintain its corrosion resistance and mechanical properties for the design life of the Yucca Mountain repository.

INTRODUCTION

Long term storage of high level radioactive waste requires reliable materials of construction. Selected materials of construction must be able to safely contain the radioactive waste and do so for long periods of time. In the United States of America, a waste repository is currently under development at the Yucca Mountain site in the state of Nevada. This repository is being designed to safely contain the radioactive waste for a period of 10,000 years. One key element of this design is the waste canister used to enclose the radioactive waste. Figure 1 shows a conceptual drawing of several of the various canister types emplaced inside of a tunnel at the Yucca Mountain site.

The canisters must have the proper strength and corrosion resistance to withstand the conditions in the waste repository at Yucca Mountain. In addition, the properties of the canister materials must remain stable over the 10,000-year design life. The high level nuclear fuel inside the waste canisters will produce heat. This heat in turn may cause metallurgical changes to occur in the canister materials over time. The ability of a material to resist metallurgical changes, called thermal stability, is a key issue for long-term integrity of the canister materials.

The temperatures at the surface of the waste canisters may exceed 200°C at first, and then slowly decay to temperatures of around 96°C within about 1,500 years (1). While temperatures of below 250°C are expected for the first 1,000 years, design guidelines use a maximum upper temperature limit of 350°C (2). Although other methods for keeping the surface temperature of the canisters below the boiling point of water are being considered, these cannot currently be relied upon. Therefore, selected materials must be able to maintain strength and corrosion resistance after long-term aging at temperatures up to 350°C.
C-22 alloy (UNS 06022) has been selected for use in the waste canister design for the Yucca Mountain Project. This material has been successfully used for chemical process, seawater service, and other demanding applications where corrosion, strength, and thermal stability are issues. However, additional data on this material were needed to predict its performance over the 10,000-year repository design life. Therefore, an extensive program was undertaken to reliably predict the long-term performance of the C-22 alloy material.

**EXPERIMENTAL APPROACH**

Samples of C-22 alloy were exposed at temperatures of 260°C to 800°C for times of up to 40,000 hours. Aging was performed in air and the samples were rapidly air-cooled, once removed from the furnace. The materials used in this study were taken from either 6.35 mm or 12.7 mm thick plate. Four different heats of material were included in this study; the chemistry of these materials is listed in Table I.
Specimens were examined for changes in microstructure, mechanical properties, and corrosion resistance. Microstructural changes were evaluated using a standard light metallograph and also using a scanning electron microscope (SEM) and transmission electron microscope (TEM). Changes in mechanical properties were determined using tensile tests (ASTM E 8) and impact energy measurements (ASTM E 23). Changes in corrosion resistance were measured using standard ASTM G 28 A tests (boiling solution of 50% H2SO4 + 42 g/l of Fe2(SO4)3) and immersion tests in boiling 2.5% HCl and 10% NaOH solutions (according to ASTM E 31). Electrochemical impedance tests were carried out according to ASTM G 106 in simulated J-13 water at 95°C. J-13 water is the designation given to the well at the Yucca Mountain site where water samples have been taken.

RESULTS & DISCUSSION

The C-22 alloy is produced in the mill annealed (MA) condition and is a metastable solid solution that has a face centered cubic (FCC) crystal structure know as gamma (γ) phase. As the C-22 alloy material is aged at various temperatures, there are several microstructural changes that can occur. These include the formation of topologically closed pack (TCP) structures, such as μ and P phases, which are know to reduce the ductility of the alloy. Secondary carbides can precipitate and grow during thermal exposure as well as the formation of a long range ordered (LRO) structure.

Microstructural changes can cause changes in the properties of a material and C-22 alloy is no exception. The extent that thermal exposure will change properties is different for each material.
The C-22 alloy has been selected for use in this application because of its excellent corrosion properties as well as its thermal stability. Microstructural changes were observed to occur for the C-22 alloy as follows (2-4):

- At 343°C and below there were no new phases formed at aging times up to 40,000 hours.
- At 427°C there was evidence of long range ordering after 30,000 hours.
- At 593°C and above there was precipitation of carbides, μ and P phases.

It should be noted that the formation of potentially harmful phases only occurs at temperatures above the maximum design temperature of 350°C. While the potentially harmful phases only occur at higher temperatures for exposure times up to 40,000 hours, a concern has been raised about their formation at lower temperatures too, given enough time. Therefore, attempts have been made to extrapolate the high temperature data to predict when these potentially harmful phases might occur at lower temperatures and longer exposure times.

The changes in impact strength as a function of aging temperature and time are shown in Figure 2. Figure 2 shows that aging will cause a decrease in toughness of the C-22 alloy material (similar to other corrosion resistant materials). This decrease in toughness corresponds to a loss of ductility of the material. It is noted that higher aging temperatures cause the toughness of the material to degrade at shorter exposure times. It should also be noted that there was no loss in impact energy for C-22 alloy material aged at 427°C, 343°C, and 260°C for times up to 40,000 hours. This data is not shown in Figure 2.

Another measure of ductility is the reduction in cross sectional area produced during a tensile test. Data from these tests mirror the results of the impact energy measurements. Loss of ductility occurs at shorter times for higher exposure temperature and very little ductility loss can be seen for temperatures of 427°C and below.

Since there will be welds in the waste package, the impact of aging on welds is also a concern. The loss of ductility in aged weld samples is similar to the response of the base metal (5). There was only a slight decrease in ductility for welded samples aged at 427°C and no loss in ductility at lower temperatures. The as welded samples contained some small amount of μ phase in the welds. This μ phase formed during cooling of the weld from the high temperatures caused by the welding process. The amount of μ phase in the welds did grow during aging of the samples at temperatures of 427°C and higher.
Fig. 2. Impact Energy of C-22 Alloy as a Function of Aging Time at Various Temperatures.

The changes in corrosion rates in ASTM G28A solution as a function of aging temperature and time is shown in Figure 3. The corrosion rates of the C-22 alloy increase when the material has been aged at higher temperatures (again, similar to other corrosion alloys). The higher temperatures will cause corrosion rates to increase at shorter exposure times. At temperatures of 538°C and below, there is essentially no change in corrosion behavior caused by thermal aging at times up to 40,000 hours. Nearly identical results were produced when using other corrosion tests, such as ASTM G28 B (boiling solution of 23% H$_2$SO$_4$ + 1% HCl + 1% FeCl$_3$ + 1% CuCl$_3$), boiling 2.5% HCl, and boiling 10% HCl. Corrosion tests have also been carried out in 10% NaOH and also simulated J13 well water; there was no effect of aging upon corrosion in these last two test solutions. J13 well water becomes caustic when concentrated; a concentration mechanism would be the boiling of water or alternate wetting and drying that would occur when ground water contacts a hot waste package. It is interesting to note that thermal aging does not seem to affect corrosion in J13 well water or caustic conditions that are the most likely environmental conditions to which the waste canister material will be exposed.
Similar results were also seen for welded samples. At aging temperatures above 482°C, corrosion rates increase with aging time for welded samples. At temperatures of 482°C and lower there is no effect of long term aging on corrosion of the C-22 alloy welds (5). Once again, an increase in the amount of $\gamma_1$ phase in the welds corresponds to an increase in corrosion rates.

In order to predict the effect that longer term aging at lower temperature might have upon the C-22 alloy, an extrapolation of data is necessary. The changes in impact energy and corrosion rates are shown on an Arrhenius plot in Figure 4. This figure shows that the changes in impact energy and corrosion rates that occur during thermal aging have very similar activation energies (i.e. similar slopes). It is believed that the changes in both of these parameters are rooted in the same microstructural feature: the precipitation of $\mu$ phase.

Should the precipitation reaction responsible for changes in material properties continue to occur at lower temperatures, the time to initiate these changes can be seen in Figure 4. Extrapolation of the curves in Figure 4 shows that, even after 10,000 years, there will be no measurable loss in either corrosion properties or in impact properties at temperatures below 350°C. This clearly demonstrates that the C-22 alloy will maintain its corrosion resistance and mechanical properties for the entire life of the waste repository at Yucca Mountain.
Fig. 4. Arrhenius Plot of Impact Energy and Corrosion Rates That Shows the Similarity of Activation Energy for Both.

CONCLUSIONS

Conclusions of this work are:

- Thermal aging at temperatures above 538°C can produce microstructural changes that can affect the mechanical and corrosion properties of C-22 alloy.
- Aging at temperatures of 427°C and below at times up to 40,000 hours does not produce any measurable change in corrosion rates of the C-22 alloy base material or welds.
- Aging at 427°C and below at times up to 40,000 hours does not produce any measurable change in ductility of the C-22 alloy base material and only a slight decrease in ductility of the welds. At temperatures of 343°C and below there was no effect of long term aging on the ductility of the welds.
- Extrapolation of high temperature data has been used to predict the effect of longer term aging (up to 10,000 years) on corrosion and mechanical properties, should the formation of μ continue to occur even at lower aging temperatures.
- Extrapolation of the higher temperature data clearly show that no degradation of corrosion properties or mechanical properties would be expected when aging C-22 alloy at 350°C or below for times up to 10,000 years.
- Based on this study, the C-22 alloy material will perform as required for the canister material in the Yucca Mountain project.
REFERENCES


