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SELECTION OF BARRIER METALS FOR A WASTE PACKAGE IN TUFF

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ABSTRACT

The Nevada Nuclear Waste Storage Investigations (NNWSI) project under the Civilian Radioactive Waste Management Program is planning a repository at Yucca Mountain at the Nevada Test Site for isolation of high-level nuclear waste. Lawrence Livermore National Laboratory is developing designs for an engineered barrier system containing several barriers such as the waste form, a canister and/or an overpack, packing, and near field host rock. In this paper we address the selection of metal containment barriers.

SUMMARY

We have selected a few candidate metals for conceptual design of canisters and overpacks, and for use in corrosion tests under repository conditions. Important materials properties data, reflecting engineering design requirements for potential candidate materials were developed. The metals that were initially considered fall into the following categories: stainless steels--austenitic, ferritic, and duplex; high-nickel alloys, titanium alloys, zirconium alloys, copper-nickel alloys, low-carbon steels, and cast irons. These metals are all commercially available.

Our procedure was to determine and evaluate the engineering properties considered to be important in meeting design requirements cost-effectively--corrosion resistance, tensile strength, weldability, etc. Four general categories were considered:

- General and local corrosion resistance
- Fabrication costs
- Required mechanical properties
- Weldability

From this analysis we selected four metals for canister and overpack materials, and one for hole liners:

1. AISI 304L stainless steel.
2. AISI 321 stainless steel.
3. AISI 316L stainless steel.
4. Incoloy 825 nickel-base alloy.
5. AISI 1020 carbon steel (for horizontal borehole liners).

The reference canister and overpack metal is AISI 304L stainless steel, but alternative metals will also be considered for two reasons: (1) to provide a replacement material until the reference material is confirmed by testing under site specific conditions; (2) to provide comparative data to support the choice of AISI 304L stainless steel as the reference material during the regulatory review.

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WASTE PACKAGE CONCEPTUAL DESIGNS

We are designing waste packages to meet the Final Rule, NRC 10CFR Part 60 and derivative requirements [1,3]. To comply with these we have developed design requirements (Table I). The waste package designs considered in the evaluation and selection of metals for canisters, overpacks and liners are given below [2,3]:

1.0 Reference designs emplaced in vertical boreholes with no liner and no packing.
    1.1 Defense high-level waste (DHLW): emplacement of 61-cm diam 304L pour canister, 1-cm thick.
    1.2 Commercial high-level waste (CHLW): emplacement of 32-cm diam 304L pour canister, 1-cm thick.
    1.3 Spent fuel: emplacement of consolidated spent fuel rods in a 304L canister, 1-cm thick.
    These dimensions are assumed by LLNL at this time for conceptual design purposes only.
2.0 Alternative designs for vertical boreholes.
    2.1 Alternative metals to 304L for canisters and overpacks.
    2.2 Overpacks for DHLW and CHLW for those canisters which do not meet acceptance criteria when received at the NNWSI repository.
    2.3 Use of packing for spent fuel canisters if the engineered system does not meet the release rate requirement.
3.0 Horizontal emplacement of waste packages within steel borehole liners.

TABLE I. The LLNL design requirements derived from NRC 10CFR60.

Waste packages shall be designed to:

1. Contain the waste for 300 to 1000 years.
2. Maintain a release rate less than 1 part in 100,000 per year of radionuclide inventory present at the end of the containment period (300 years minimum).
3. Be retrievable for 50 years after emplacement of the first waste package.
4. Meet nuclear criticality standards, e.g., not exceed an effective multiplication factor (K_{eff}) of 0.95.
5. Not exceed temperature limits of the waste forms, which are 773 K (500°C) for DHLW glass, 673 K (400°C) for CHLW glass, and 623 K (350°C) for spent fuel (this is necessary to meet the 1 part in 100,000 per year requirement).
6. Not leak radioactive material in excess of applicable federal and state standards after a drop test of two times waste package length onto an unyielding surface, at the minimum anticipated handling temperature.
7. Not leak radioactive material in excess of applicable federal and state standards after sustaining a 1073 K (800°C), 30-min fire test.
8. Not leak radioactive material in excess of applicable federal and state standards during or after transportation, handling, emplacement, retrieval and expected seismic loads. Further, these loads must not compromise long-term performance.
9. Retain legible, externally labeled identification up to and including retrieval.
10. Meet federal regulatory requirements for transportation of high level nuclear waste (DHLW and CHLW pour canisters).
11. Meet requirements with considerations for cost-effectiveness, including direct package costs and related repository system costs through the operational period.

EMPLACEMENT ENVIRONMENT

The NNWSI project has selected the Topopah Spring Member of the Paintbrush Tuff as the target horizon for a repository sited at Yucca Mountain. The repository will be located in a welded portion of the tuff unit and will lie approximately 350 to 400 meters below the surface level. The static water level is over 100 meters below the repository level. The waste canisters will not be submerged in water, but will be subjected to constant contact with water vapor and to intermittent contact with about 8 mm/yr of seeping liquid water [3]. The pressure exerted on the canisters by the environment will be approximately 1 atmosphere with no hydrostatic pressure because there is not a continuum of water above or around the canisters. The gaseous environment to which the canister is exposed will be of air plus water vapor when the temperature is more than 95-100°C.

Wetted corrosion of the canister or overpack will begin after the temperature has dropped to less than 95-100°C. This is because liquid water cannot exist in the unsaturated zone (vadose zone) at temperatures higher than 95-100°C, the 1 atmosphere boiling point of water. Calculations of thermal history for canisters and overpacks show that for DHLW, the temperature drops below 100°C in about 60 years after emplacement and for CHLW, temperature will drop below 100°C approximately 180 years after waste emplacement. For spent fuel packages, the canister temperature will remain 100°C or greater for approximately 1000 years after emplacement. The unsaturated zone just above the water table and atmosphere of the repository will be mildly oxidizing, and there will be a decaying gamma flux starting at up to 1.1×10^5 rem/hr for commercial high-level waste [2].

NOMINATION OF CANDIDATE METALS

A list of 17 candidate metals which potentially will meet our design requirements is given in Table II. These metal alloys are of three types: (1) iron-base alloys with a ferritic structure; (2) iron-base to nickel-base alloys with an austenitic structure; and (3) copper, titanium, and zirconium-base alloys.

The list of metals will be screened to yield the five top-contenders.

1. Iron-base alloys with a ferritic structure

The ferritic metals considered for this group are low-carbon steels, that may have a high corrosion rate in the anticipated oxidizing environment of the repository. The redeeming properties of these steels are: lowest overall unit cost; acceptable strength at room temperature and 800°C; and good weldability. We considered two low-carbon steels, AISI 1020 steel and ASTM A537B steel. Carbon steel is the reference metal for horizontal borehole liners, based on its low cost and projected survival during the retrieval period.

Alloying carbon steels with chromium and molybdenum increases corrosion resistance under oxidizing conditions. The ferritic alloy steels under consideration are expected to be resistant to attack by pitting and crevice corrosion as well as to stress corrosion cracking. The shortcomings of this group are low fracture toughness values as well as poor weldability. The ferritic alloy steels considered are AISI 409 Ti stabilized stainless steel and 26 Cr - 1 Mo stainless steel.

2. Iron-base to nickel-base alloys with an austenitic structure

The NNWSI reference canister and overpack metal, which also is the reference for the DHLW and CHLW pour-canisters, is AISI 304L stainless

TABLE II. Candidate metals for overpacks.

Commercial material designation	Chemical composition (wt%)
1. AISI 1020 cs (UNS G10200)	C .18-.23, Mn .3-.6, P .04 max, S .05 max
2. ASTM A537B cs	C .24 max, Mn .7-1.35, P .035 max, S .04 max, Si .15-.5, Cr .25 max, Ni .25 max, Mo .08 max, Cu .35 max
3. AISI 409 ss (UNS S40900)	C .08 max, Cr 10.5-11.75, Mn 1.0 max, Ni 0.50 max, P .04 max, S .045 max, Si 1.0 max, Ti 6XC min -0.75 max
4. 26 Cr - 1 Mo ss (UNS S44626)	C .06 max, Cr 25.-27., Cu .2 max, Mn .4 max, Mo .75-1.50, N .04 max, Ni .05 max, P .04 max, S .02 max, Si .75 max, Ti 0.20-1.00, Other Ti 7X (C+N) min
5. AISI 304L ss (UNS S30403)	C 0.030 max, Cr 18.00-20.00, Mn 2.00 max, Ni 8.00-12.00, P 0.045 max, S 0.030 max, Si 1.00 max
6. AISI 321 ss (UNS S32100)	C 0.08 max, Cr 17.00-19.00, Mn 2.00 max, Ni 9.00-12.00, P 0.045 max, S 0.030 max, Si 1.00 max, Ti 5X C min
7. AISI 316L ss (UNS S31603)	C 0.030 max, Cr 16.00-18.00, Mn 2.00 max, Mo 2.00-3.00, Ni 10.00-14.00, P 0.045 max, S 0.030 max, Si 1.00 max
8. AISI 317L ss (UNS S31703)	C 0.030 max, Cr 18.00-20.00, Mn 2.00 max, Mo 3.00-4.00, Ni 11.00-15.00, P 0.045 max, S 0.030 max, Si 1.00 max
9. Nitronic 33 ss (UNS S24000)	C 0.08 max, Cr 17.00-19.00, Mn 11.50-14.50, N 0.02-0.40, Ni 2.50-3.75, P 0.060 max, S 0.030 max, Si 1.00 max
10. JS 700 ss (UNS N08700)	C .04 max, Ni 24.0-26.0, Cr 19.0-23.0, Mo 4.3-5.0, Nb 8X C min-.04 max, Si 1.0 max, Mn 2.0 max, P .04 max, S .03 max, Cu .5 max
11. Ferralium 255 ss (UNS S32550)	C .04 max, Cr 24.0-27.0, Mo 2.0-4.0, Ni 4.5-6.5, Si 1.0 max, Mn 1.5 max, N .10-.25, Cu 1.5-2.5
12. Incoloy 825 (UNS N08825)	Al 0.2 max, C 0.05 max, Cr 19.5-23.5, Cu 1.5-3.0, Fe bal, Mn 1.0 max, Mo 2.5-3.5, Ni 38.0-46.0, S 0.03 max, Si 0.5 max, Ti 0.6-1.2
13. Inconel 625 (UNS N06625)	Al 0.40 max, C 0.10 max, Nb 3.15-4.15, Cr 20.0-23.0, Fe 5.0 max, Mn 0.50 max, Mo 8.0-10.0, Ni bal, P 0.015 max, S 0.015 max, Si 0.50 max, Ti 0.40 max
14. Ti Grade 2 (UNS R50400)	C 0.10 max, H 0.015 max, Fe 0.30 max, N 0.03 max, O 0.25 max, Ti Rem

15.	Ti Grade 12	N .03 max, C .08 max, H .015 max, Fe .3 max, O .25 max, Mo .2-.4, Ni .6-.9, Ti Rem
16.	Zr 702 (UNS R60702)	C 0.05 max, H 0.005 max, Hf 4.5 max, N 0.025 max, Other Zr + Hf 99.2 min, Fe + Cr 0.2 max
17.	Cupronickel 70/30 (UNS C71590)	Cu 67.0 min, Ni 29.0-33.0, +...

steel. The engineering properties of 304L stainless steel rank very well with the exception of susceptibility to localized corrosion and to stress corrosion cracking. If intergranular stress corrosion cracking is excessive for 304L stainless steel in the Topopah Spring environment, other stabilized austenitic stainless steels, such as 321 stainless steel, or nickel-base alloys, such as Incoloy 825, are appropriate choices. If transgranular stress corrosion cracking is a problem for the austenitic stainless steels, an alloy with greater than 20% nickel content [4,5], such as Incoloy 825, is appropriate.

If pitting and/or crevice corrosion attack is excessive in 304L stainless steel, then an alloy with increased molybdenum, e.g., AISI 316L and AISI 317L stainless steels, or Incoloy 825, will increase the resistance to these forms of corrosion. All of the 300 series stainless steels may be specified with the extra-low carbon (.02 max) modification to avoid sensitization in the high-temperature glass pouring process, and in the final top-cap weld on canisters and overpacks.

3. Copper, Titanium, and Zirconium-base alloys

The titanium alloys are expected to be very resistant to conditions that may occur in a strong field of gamma radiation. These alloys are also very resistant to localized forms of corrosion, but lose most of their mechanical strength at 800°C. When compared to the other metals, cupronickel 70/30 has competitive strength and weldability properties, but is vulnerable to corrosion by nitric acid and other oxidizing species in a radiolyzed air-water environment. Zirconium alloys have proven performance in aqueous, radiolyzed environments, but exhibit low mechanical strengths at 800°C as well as marginal fracture toughness values at -18°C, and are the most costly of the candidate metals considered.

1. Corrosion Data

There is abundant evidence in the literature [6] that when moist air is irradiated with ionizing radiation, nitric acid will form. In radiation corrosion experiments with moist air [7-10], it has generally been found that metals known to be vulnerable to corrosion by nitric acid are also corroded in irradiated, moist air. For conditions in the tuff repository, the relative increases in corrosion rates would probably be significant for copper-based alloys. Makepeace [10] reported a 6-month irradiation experiment which indicated that certain austenitic stainless steels and nickel-based alloys had very low corrosion rates in moist air irradiated with a gamma-ray flux of 5 to 6 megarads per hour at ambient temperature. Under the same conditions, copper samples showed higher corrosion rates. Although this experiment was carried out at a higher dose rate, a lower temperature, and for much shorter times than are of interest for the tuff repository, the results are indicative of the type of corrosion behavior to be expected under irradiation conditions.

Presently no long-term underground corrosion data exist for our candidate metals in an environment exactly like that at Yucca

Mountain [13]. However, underground corrosion data for a few of the candidate metals were found for Chino silt loam for a 14-year test [11]. Table III lists the significant attributes of both environments and presents available data. The data indicate the following for the Chino silt loam environment:

- A Mo-containing austenitic stainless steel (i.e., type 316) performed better than an ordinary Ni-Cr austenitic stainless steel (i.e., type 304).
- Ferritic stainless steels (i.e., types 430 and 410) did not perform as well as 304 stainless steel.

Performance was determined by general corrosion and pitting corrosion rates on coupons of these alloys buried in soil. Relationships between the corrosivity of soils and factors such as soil resistivity, pH, aeration, and the corrosion products formed, have been suggested in the literature [11,12].

Although major differences are shown between the "Chino silt loam" environment and the Topopah Spring tuff of Yucca Mountain environment, there are also some similarities, and use of these data represent estimates for long-term underground conditions until site-specific data for the repository environment become available.

2. Material and Fabrication Costs

Rolled and welded pipe manufacturing processes are representative of the kind of fabrication involved in manufacturing canisters and overpacks. Diameters of 0.3 m to 0.91 m represent the upper limit of canisters and overpack diameters that we contemplate in our designs, so

TABLE III. Underground environments.

	Topopah Spring tuff [2]	Chino silt loam [11] (14-yr data)
Chemical concentrations of water extract (ppm)	SiO_2-61.0, Na-51.0, K-4.9, Ba-0.003, Ca-14.0, Mg-2.1, Fe-0.04, Al-0.03, F-2.2, Cl-7.5, NO_3-5.6, Li-0.05, Sr-0.05, SO_4-22.0, PO_4-0.12, HCO_3-120.0	Na+K-76.5, Ca-124, Mg-22, HCO_3-13, Cl-60.5, SO_4-169
Water	8 mm/yr (net)	386 mm/yr (average rainfall furnished by the U.S. Weather Bureau)
Resistivity	high	low
pH	∿ neutral	∿ neutral
Redox	oxidizing	oxidizing
Air-pore space	17%	16%
% moisture	14%	26%
Atmosphere	air-water/film-steam	air-water/film
Temperature	29-250°C	10°C-28°C
Radiation field	gamma, 1.1×10^5 rem/hr max	background
Areal thermal loading	50 kW/acre	solar
Corrosion products	scale	scale
Pitting corrosion data		
AISI 304 s. stl.	TBD (to be determined)	1.1 mpy (28 μm/yr)
AISI 316 s. stl.	TBD	4×10^{-5} mpy (10^{-3} μm/yr)
AISI 430 s. stl.	TBD	4.4 mpy (112 μm/yr)
AISI 410 s. stl.	TBD	4.4 mpy (112 μm/yr)
Internal drainage	TBD	good

we used the cost of 0.91 m (36 in.) diameter by 12.7 mm (1/2 in.) wall welded pipe as a measure. These costs were obtained by telephone contact with commercial fabricators. The costs of alternative fabrication processes such as extrusion and centrifugal casting were also considered, but were rejected due to high cost or failure to meet our minimum design requirements.

3. Mechanical Properties

Mechanical properties of interest include fracture toughness at 255 K (-18°C), elongation, nil-ductility temperature, tensile strength, and yield strength at 1073 K (800°C). Fracture toughness is dictated by the requirement that the waste package survive a drop test from a height of two times the package length without leaking. During transportation and at Yucca Mountain, minimum handling temperatures are roughly 255 K (-18°C). The ductility (elongation) is also important to the drop test requirement. The fracture toughness of certain alloys exhibits significant variations with changes in temperature.

The ultimate tensile strength is a measure of the stress which will lead to fracture of the material. The margin between the yield strength and the ultimate strength is a measure of the degree to which stretching and bending rather than fracture takes place after the yield strength is exceeded. Actual designs will load the material to a stress below the yield strength including a factor of safety. The yield strength at 1073 K (800°C) is related to the requirement of surviving a 1073 K (800°C), 30-min fire test without leaking.

4. Weldability

The waste package containment barrier will have several welded joints. The final weld of the top-cap to the main overpack and canister bodies will be done remotely. Possible welding problems were identified by yes (1) or no (0) binary evaluation of the following characteristics: preheat requirements, special interpass temperature, postheat requirements, special welding atmosphere, low weld toughness, nonstandard welding process, nonstandard nondestructive evaluation process, special cleanliness during fit-up, and not economical relative to 304 stainless steel. The overall weldability also includes all dimensional and mechanical property requirements of the weld and heat-affected zone.

RESULTS AND RECOMMENDATIONS

Analysis of the data resulted in selection of five metals out of the seventeen candidates. Table IV scores and ranks the candidates.

The following summarizes the metals chosen as top-ranked in satisfying all of the design requirements for canisters and overpacks.

1. AISI 304L Stainless Steel - a low carbon, general-purpose austenitic stainless steel. We will further specify a premium grade with a carbon content less than 0.02%·C if experimental results and analysis indicate that chromium carbide precipitation (sensitization) will occur during welding and glass pouring. This is designated the reference metal.

2. AISI 321 Stainless Steel - a general-purpose, austenitic stainless steel with a titanium addition for stabilization of the carbon, thus preventing the formation of chromium-carbides (sensitization) during welding and glass pouring, as well as over long periods of time, at low temperatures (100-300°C) (alternative).

3. AISI 316L Stainless Steel - a low carbon, austenitic stainless steel with the addition of 2-3% molybdenum for more resistance to pitting corrosion than type 304L. We will further specify a premium grade with a

TABLE IV. Ranking summary for candidate metals.

Material designation or composition	Corrosion resistance	Mechanical properties	Weld-ability	Cost	Score	Rank[b]
AISI 1020 steel	0[a]	1	2	2	5	3
A537 steel	0	2	1	2	5	3
409 st. steel	1	1	0	1	3	3
26 Cr - 1 Mo steel	1	1	0	0	2	3
304L st. steel	1	2	2	2	7	1
321 st. steel	1	2	2	2	7	1
316L st. steel	1	2	2	2	7	1
317L st. steel	1	2	2	1	6	2
Nitronic 33	1	2	2	1	6	2
JS 700	2	2	0	1	5	3
Ferralium 255	1	1	1	1	4	3
Incoloy 825	2	2	2	1	7	1
Inconel 625	2	2	2	0	6	2
Ti Code 2	2	0	1	0	3	3
Ti Code 12	2	0	0	0	2	3
Zr 702	2	0	0	0	2	3
Cu - Ni 70/30	0	2	1	1	4	3

[a] 0 = some disadvantages; 1 = suitable; 2 = superior.
[b] 1 = highest; 3 = lowest.

carbon content less than 0.02% C if experimental results and analysis indicate that chromium carbide precipitation (sensitization) will occur during welding and glass pouring (alternative).

4. Incoloy 825 - a nickel-iron-chromium-molybdenum-copper austenitic alloy designed for extremely corrosive environments. This alloy is stabilized with titanium to resist intergranular corrosion and intergranular stress corrosion cracking. The nickel content makes it very resistant to transgranular stress corrosion cracking. The molybdenum and copper give this alloy resistance to pitting and crevice corrosion. The high chromium content gives it resistance to various types of oxidizing environments (alternative).

5. AISI 1020 Steel - a low carbon, general-purpose steel for the reference metal, for horizontal borehole liners, appropriate for a 50-year retrieval period.

CONCLUSION

Our analysis indicates five metals that best satisfy the requirements for disposal of high-level waste at the NNWSI proposed repository at Yucca Mountain. Testing is in progress on these metals for further development of the waste package design for the unsaturated zone. The reference canister and overpack metal is AISI 304L stainless steel, and the primary alternative metals are AISI 321, AISI 316L, and Incoloy 825. For borehole liners, 1020 carbon steel has been chosen as the reference metal.

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