FEASIBILITY ASSESSMENT OF COPPER-BASE

WASTE PACKAGE CONTAINER MATERIALS IN NUCLEAR WASTE REPOSITORIES

SITED IN BASALT AND TUFF

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ABSTRACT

In early 1984, the United States Department of Energy Office of Civilian Radioactive Waste Management (DOE-OCRWM) established a two-year program (FY 1985 and FY 1986) to evaluate the use of copper and copper alloys in basalt and tuff repository environments in accordance with Congressional directive. The Basalt Waste Isolation Project (BWIP) and the Nevada Nuclear Waste Storage Investigation (NNWSI) Project concluded that copper-base materials are feasible as candidate container materials in a repository sited in a basalt or tuff environment.

The feasibility of using copper materials in containers was qualitatively assessed using the following criteria: 1) container design and development; 2) preclosure safety (e.g., fabrication or emplacement risks); 3) repository interfaces (e.g., handling of containers); 4) retrievability considerations; 5) containment (mainly corrosion considerations); 6) radionuclide release (container/waste interactions); and 7) cost and availability. Weighting factors were not used and no comparison to other candidate disposal container materials was made.

This paper details the results of testing, literature reviews, and evaluations that were performed for each of the seven criteria on each of the three conceptual container designs. The designs were as follows: A thick-walled Cupronickel 90-10 pressure vessel (BWIP); a copper monolith made by the HIP (hot isostatic pressing) process (BWIP); and a thin-walled aluminum bronze or Cupronickel 70-30 pressure vessel (NNWSI Project). A brief discussion of future plans to evaluate copper-base materials is presented for the BWIP and NNWSI Project.

BACKGROUND

To satisfy the "substantially complete containment" requirements of 10 CFR 60, the Office of Civilian Radioactive Waste Management (OCRWM) within the Department of Energy (DOE) has focused on the evaluation of metals and alloys for use in a disposal container. In early 1984, in accord with Congressional intent (1), the DOE established a two-year program (FY 1985 and FY 1986), now completed, to evaluate the use of copper and copper alloys in basalt and tuff repository environments. The DOE consulted with the International Copper Research Association (INCRA) and the Copper Development Association (CDA) on the formulation of the programs in basalt and tuff, including the selection of copper and copper alloys to be evaluated. The CDA and INCRA performed some of the tasks outlined in the study.

The feasibility of using copper materials in containers was assessed qualitatively with respect to the following evaluation criteria: 1) container design and development; 2) preclosure safety (e.g., fabrication or emplacement risks); 3) repository interfaces (e.g., handling of containers); 4) retrievability considerations; 5) containment (mainly corrosion considerations); 6) radionuclide release (container/ waste and container/cladding interactions); and 7) cost and availability. Weighting factors were not used and no comparison to other candidate disposal container materials was made.

The Basalt Waste Isolation Project (BWIP) and the Nevada Nuclear Waste Storage Investigations (NNWSI) Project completed their two-year programs and concluded that copper-base materials are feasible as candidates in their container materials testing programs. Results of FY 1985 efforts and FY 1986 plans were summarized in a status report to Congress (2) and detailed in the BWIP and NNWSI project status reports (3, 4). This paper provides the conclusions as reported to Congress (5) of the two year copper assessment program. More detailed, project specific feasibility assessment reports were prepared by the BWIP and NNWSI Project (6, 7).

DESCRIPTION OF THE BWIP FY 1985 AND FY 1986 COPPER EVALUATION PROGRAM

Expected Conditions During Container Lifetime in a Basalt Repository

The preferred basalt repository horizon is the Cohassett flow located at a depth of approximately 900 m below the surface within the Grande Ronde basalt formation. The environmental conditions expected in the vicinity of the waste package in the basalt repository are an oxidizing water vapor and air environment during the operating period (50-100 years) and sometime into the postclosure period with a gradual change into a reducing aqueous environment. In addition, during the post-closure period, the waste package will be subjected to a hydrostatic pressure of approximately 9.1 MPa after resaturation. The natural environment of the repository will be further perturbed after emplacement of the waste package primarily due to the generation of heat from radioactive decay.

Identification of Candidate Materials

At the end of FY 1984, the BWIP developed a test plan for evaluating copper and copper-base alloys as alternative container materials for an HLW package for disposal at a potential repository site in basalt rock. Short-term testing on Cupronickel 90-10 had begun earlier (FY 1982) as a result of studies on the stability of copper deposits in native basalts. The testing program on copper-base container materials was expanded to its present scope in FY 1985 in response to the Congressional directive. The two candidate materials selected at the beginning of FY 1985 for detailed testing were oxygen-free high conductivity (OFHC) copper and Cupronickel 90-10. Phosphorus-deoxidized (PDO) copper was also evaluated in a few selected tests to determine if the uniform corrosion behavior of copper depends on minor alloy element variations.

BWIP FEASIBILITY CRITERION 1: CONTAINER DEVELOPMENT AND DESIGN

Container Designs

Two basic container design concepts were considered by the BWIP. In both designs the container is surrounded by a packing material of 75% (by weight) crushed basalt with 25% bentonite, and the packing and container are encased in a thin metal shell to facilitate emplacement.

The first (reference) design is a pressure vessel type design employing a Cupronickel 90-10 cylinder enclosing a waste form of spent fuel or vitrified high level waste (HLW). The minimum wall thicknesses of the containers are conservatively sized to withstand hydrostatic pressure in the repository. An additional thickness is added as an allowance for uniform corrosion. The container thicknesses are further increased where necessary to ensure that possible enhancement of corrosion rates by radiolysis would be insignificant. The resulting wall thicknesses were 9.4 cm and 13.8 cm for consolidated spent fuel and HLW forms, respectively. A pressure vessel design concept using OFHC copper was not considered feasible due to the low strength of this material at the expected repository temperatures and also due to lack of mechanical property data at elevated temperatures.

The second (alternate) design is a solid monolith type (similar to that under consideration in Sweden) and employs pure copper (although Cupronickel 90-10 could also be used). The design consists of 4 intact pressurized water reactor (PWR) spent fuel assemblies encapsulated in a fully densified solid matrix of copper formed by the hot isostatic pressing (HIP) process from copper powder in a copper casing. The monolith concept also met the criterion for limitation of radiolysis-enhanced corrosion.

Radiation Shielding Analysis

Preliminary calculations of gamma radiation levels expected in the waste package during the containment period were performed. The dose rates were calculated for 33,000 Mwd/MTU fuel 10 years out of reactor at time of emplacement. Gamma dose rates at the container surface for the nominal wall thickness were found to be as follows: approximately 80 rad/h for consolidated spent fuel (9.4 cm thick), approximately 1 rad/h for intact spent fuel (adjacent to an assembly corner) (15 cm thick), and approximately 2 rad/h for HLW (13.8 cm thick). In the case of high burnup fuel, (60,000 Mwd) the dose rate increased by a fuel factor of approximately 2.

Fabrication, Closure, and Non-Destructive Examination Techniques for Copper-Based Containers

The CDA assessed the adequacy of currently available technology to fabricate, close, and inspect copper containers. CDA concluded that the two BWIP designs (and the NNWSI Project thin-walled design) could be made utilizing currently available technology. The BWIP reference fabrication method of centrifugal casting was concluded to be well-suited for production of a Cupronickel pressure vessel container. Closure welds could be made with electron beam methods and inspection could be performed by radiography or ultrasonic inspection. The HIP process was presented as a feasible process for production of the alternate concept of a monolith of either pure copper or copper alloys. However concern exists relative to the need for high throughput production of the container using the HIP process in a remote-handling operation.

Some developmental work would be needed in the following areas: automatic welding, extrusion of containers of the desired size, process parameters for the HIP process, and remote handling technology. Work of this nature is needed to some extent no matter what the choice of container material is.

Container Design and Development Feasibility Evaluation (Summary)

Current design studies show that Cupronickel 90-10 is feasible for use as a pressure vessel type waste container. More elevated temperature mechanical property data are necessary to permit confirmation of design assumptions about structural stability and resistance to creep deformation. The monolith concept is feasible for OFHC copper, PDO copper, or Cupronickel 90-10.

BWIP FEASIBILITY CRITERION 2: PRE-CLOSURE SAFETY

Preclosure safety refers to accident probabilities and consequences during container fabrication and emplacement. This does not appear to be a significant concern as a result of using copper-based materials in containers. Copper's ductility would probably allow it to survive accidental drops during handling. Preclosure safety is not an issue with a centrifugally cast container as such containers would be purchased from a vendor rather than cast at the repository site. The situation is different with HIP containers as they would be fabricated on site. The HIP process is widely used for non-nuclear applications in the United

States. However, the transition from non-nuclear industrial practice to hot cell operation for a process that requires high pressures (150 MPa), high temperature (550°C), and the possible use of hydrogen, could require more complicated design features and administrative control to guard against process upset or accidents.

BWIP FEASIBILITY CRITERION 3: REPOSITORY INTERFACES (HANDLING)

There are aspects of container handling that will be affected by the choice of copper materials for a container. Handling fixtures will have to be carefully designed so that ductile yielding, particularly of pure copper, will not compromise firm gripping. The weight of a Cupronickel pressure vessel container presents no more significant a challenge to handling than a steel container would. The additional weight of a monolith will present a challenge to handling, emplacement, and retrieval equipment design, although there is a potential for design to accommodate the weight. Thus, copper is feasible from a repository handling standpoint, with further studies required to ensure weight accommodation by equipment design.

BWIP FEASIBILITY CRITERION 4: RETRIEVABILITY

There is no apparent impact, other than the aspects noted above under handling, of the usage of copper containers on retrievability.

BWIP FEASIBILITY CRITERION 5: EVALUATION OF CONTAINMENT POTENTIAL

Types of Copper Degradation for Consideration in Expected Basalt Repository Environments

The BWIP program concentrated on studies of uniform corrosion, pitting corrosion, and susceptibility to environmentally-assisted cracking (slow-strain-rate studies and fracture mechanics studies). The degradation mode expected to have the highest probability of occurrence in the basalt repository environment is uniform corrosion due to reaction of the copper materials with oxygen under air/steam conditions or sulfides after the onset of anoxic conditions. Potential localized corrosion modes include pitting, crevice corrosion, intergranular corrosion, environmentally-assisted cracking (EAC), i.e., stress corrosion cracking (SCC) and hydrogen embrittlement. Selective leaching of nickel in Cupronickel 90-10 is another possible corrosion mode. SCC can be caused by the presence of ammonium and nitrite ions, but these are expected to exist in the repository environment only in trace quantities. The copper alloys are expected to be highly resistant to hydrogen embrittlement, selective leaching, and crevice corrosion.

Uniform Corrosion

Short-term tests were conducted on Cupronickel 90-10, OFHC copper, and PDO copper at 50, 100, 150, and 200°C in anoxic synthetic basalt groundwater plus packing material in low-flow autoclaves and static pressure vessels. The air/steam tests were conducted at 150, 200, 250 and 300°C in order to simulate the preclosure phase and the unsaturated postclosure phase of the containment period, while the groundwater test environment simulated the saturated postclosure phase.

At 250°C, a probable maximum temperature for the air/steam environment, the average rates after 10 months of air/steam exposure were 1.7 microns per year for Cupronickel 90-10 and 20.7 to 27.4 microns per year for PDO and OFHC copper.

At 200°C, a probable maximum temperature for groundwater exposure of containers, the corrosion rates after 10 months with autoclaves were 12.2 microns per year for Cupronickel 90-10 and 2.3 to 3.9 microns per year for the pure copper. The pressure vessel tests at 200°C yielded results of 6.5 microns per year for Cupronickel 90-10 and 6.9 to 17.5 microns per year for pure copper. The reasons for the differences between the two test types are not presently known but are under investigation.

The effect of irradiation on the corrosion of OFHC copper and Cupronickel 90-10 was studied in autoclaves at 100 and 200°C. Test conditions were similar to those described previously for uniform corrosion experiments with the addition of a gamma radiation field ranging from 100 to 10,000 R/hr. Maximum corrosion rates based on weight loss observed at 200°C after four months exposure were 16.1 microns per year for Cupronickel 90-10 and 20.7 microns per year for OFHC copper as compared with 17.1 and 19.4 microns per year respectively in the absence of radiation. Therefore, the data indicate no significant effect of radiation exceeding expected design levels on the corrosion rate of these alloys for the range of conditions investigated. No selective leaching was apparent in uniform corrosion specimens examined with optical metallography.

Pitting Corrosion

Cupronickel 90-10 and OFHC copper were tested for susceptibility to pitting by obtaining polarization curves at 50, 100, 150, and 200°C, initiating long-term corrosion potential and polarization resistance monitoring tests at 100, 150, and 200°C, and testing for pit growth rates in thin-walled specimens at 150 and 200°C. Tests were conducted in synthetic basalt groundwater with packing material under anoxic conditions. The polarization curves suggest the possibility of passive film formation for high-purity copper at 150°C and 200°C. Formation of a passive film is a potential precursor to pitting; however, no pitting was observed in post-test examination of the specimens. Long-term (up to 8000 h) monitoring tests indicate that the actual corrosion potential remained below the indicated pitting potential, as suggested by the polarization curves. No evidence of pitting in thin-walled pit growth specimens has been obtained for copper or Cupronickel 90-10 at 200°C for times up to 2100 h. Furthermore, pitting was not detected after metallurgical examination of a statistical sampling of cross-cut sections in any of the uniform corrosion test specimens in any of the test environments described in the preceding section. Although conclusions about the long-term pitting behavior of the copper-base materials cannot be made on the basis of these short-term studies, nothing detrimental to copper was indicated.

Environmentally Assisted Cracking

Slow strain rate tests in synthetic basaltic groundwater at 100 and 200°C and 1×10^{-4} and $2 \times 10^{-7} sec^{-1}$ strain rate showed no effects of the groundwater on the ductility of OFHC copper and Cupronickel 90-10. This indicates no tendency for environmentally-assisted cracking.

Observed stress relaxation behavior suggests that environmentally assisted cracking would be difficult to initiate in OFHC copper above 100°C and in Cupronickel 90-10 above 150°C as the stress levels at a crack tip required for propagation cannot be maintained at the temperatures expected in the waste package. Threshold temperatures for crack propagation are expected to be exceeded for spent fuel waste packages for 1000 y after emplacement.

Containment Feasibility Evaluation (Summary)

Corrosion testing and literature studies to date have revealed no properties that would exclude copper-base materials from consideration as a waste container material in a basalt repository. If uniform corrosion rates obtained to date in anoxic environments are linearly extrapolated to 1000 years, reasonable corrosion allowances of 1 to 2 cm are obtained. Therefore, based on the limited tests summarized here, it is feasible for copper-based containers to resist corrosion for the necessary time frame to meet regulatory requirements. However, a possible problem area is the enhancement of uniform corrosion by increased sulfide concentrations due to the microbial reduction of sulfate to sulfide. Since there are only qualitative estimates now of sulfide concentrations and enhanced uniform corrosion possible from microbial action, the magnitude of the corrosion that may result still needs to be defined. In addition, longer-term studies are underway to evaluate the susceptibility of copper-base materials to pitting, crevice corrosion, or environmentally-assisted cracking in a basalt repository environment in the presence of radiation.

BWIP FEASIBILITY CRITERION 6: EVALUATION OF EFFECTS OF COPPER ON RADIONUCLIDE RELEASE CHARACTERISTICS

Two hydrothermal experiments were completed to evaluate the influence of copper on the behavior of spent fuel and spent fuel/basalt in autoclaves containing synthetic basalt groundwater in the absence of bentonite to see if copper would adversely affect radionuclide release. The tests were run for a maximum of approximately 4000 h, at a temperature of 200°C. The preliminary solution data present no evidence of any significant reaction between copper and the spent fuel or the basalt. There were no significant differences in measured concentrations of radionuclides or major anions and cations between experiments with and without copper. Therefore, the use of copper should have no negative impact on radionuclide release subsequent to container breach based on experimental results to date. More work is planned in this area.

BWIP FEASIBILITY CRITERION 7: COST AND AVAILABILITY OF COPPER-BASED MATERIALS

Another important aspect of the feasibility of copper is cost and availability. In an extensive review by CDA, it was reported that the United States is essentially self-sufficient in copper. The U.S. supply of copper through the year 2000 will be more than adequate to meet the demand imposed by the use of copper materials in high-level nuclear waste containers. The total amount of copper needed to fabricate the required number of containers for a repository in either a basalt or tuff medium ranges from 30,000 to 100,000 metric tons over a 25-year period. On an annual basis, this is 1200-4000 metric tons per year or 0.04-0.15% of U.S. consumption.

The price of refined copper is expected to increase only slightly from the current approximate price of 65 cents per pound (1986 dollars) and is not expected to exceed 90 cents per pound through the year 2000. These analyses show copper-based materials to be cost-competitive as container materials. Forecasts of copper supply and price beyond the year 2000 are not available.

The cost of materials and fabrication of a centrifugally cast the BWIP Cupronickel 90-10 container for consolidated spent fuel was estimated to be $50,800 (in 1985 dollars); this included the complete cost of design, materials, fabrication, and shipping to the repository, as well as the emplacement shell. The thicker vitrified waste container was estimated to cost $87,000. Assessments of cost significance are difficult without employing relative measurements or comparisons. However, the container costs are not expected to impact copper container feasibility for a basalt repository.

SUMMARY OF COPPER CONTAINER FEASIBILITY EVALUATION (BWIP)

Feasibility of Using a Copper-Base Container in a Basalt Repository

It is concluded that copper is feasible for use as a waste container for a repository in basalt, based on the limited investigations conducted to date. Short-term investigations under limited test conditions have not uncovered corrosion modes or rates that are expected to lead to failure in 300 to 1000 years. More study will be required, particularly in the area of sulfide corrosion from anaerobic microbial action, before definitive answers can be provided. Indications are that copper will not have an effect on radionuclide release. The pressure vessel design can be fabricated with existing technology. Technology would need to be developed for the monolith container. Elevated temperature tensile property data will be needed to complete a pressure vessel design. Cost and availability should present no barrier to copper usage. There should be no significant impact on repository interfaces and retrievability of a copper alloy as a container material if a pressure vessel design concept is chosen. Use of a monolith container also appears to be feasible.

PLANS FOR FURTHER EVALUATION OF THE USE OF A COPPER-BASE CONTAINER IN A BASALT REPOSITORY

BWIP will continue evaluation of pure copper and Cupronickel 90-10 along with other candidate materials, A27 carbon steel and A387 low alloy steel, until a final reference material is selected. In early FY 1987, as part of the Site Characterization Plan (SCP) process, Study Plans for corrosion testing, design, and analysis will be written and implemented to describe the methodology and integration of efforts leading to a licensable waste package design. During FY 1987 and FY 1988, testing will continue in the areas of uniform corrosion, pitting, and environmentally assisted cracking. Studies on the effect of radiation on uniform and localized corrosion will be continued. Studies of microbial effects on corrosion will be carried out and recommendations for testing of such effects will be issued. Mechanistic studies to couple theoretical knowledge with empirical data will be implemented. Studies to determine the "worst case" environment probable at the container surface, from local concentration and reaction effects, will be performed and the results will be incorporated in the testing program. Further experimental work on radionuclide concentrations involving copper-based materials is also planned. Further evaluations of HIP process preclosure safety and effect of monolith container weight on retrievability will be made to assess the resulting impacts on the repository interfaces and retrievability by use of a copper monolith. Advanced conceptual designs for waste packages for a repository in basalt are currently planned to be completed in the 1989 timeframe. During subsequent design phases, it is planned that corrosion testing and analysis will be performed to a sufficient degree so as to provide a framework for the final selection of a container material.

DESCRIPTION OF THE NNWSI PROJECT FY 1985 AND FY 1986 COPPER EVALUATION PROGRAM

Expected Conditions During Container Lifetime in a Tuff Repository

The repository horizon is in a layer of welded devitrified tuff in (Topopah Spring Member) Yucca Mountain at least 200 m above the static water table (at a depth of approximately 365 m). The effective pressure in the repository will be atmospheric. The porous nature of the rock implies that air will be present and that the limited amount of vadose water will be air-saturated. (The projected water flux at the repository horizon is less than 0.5 mm/y downward. Rainfall at Yucca Mountain is less than 150 mm/y and most of the water evaporates before it can enter the ground.) Heat and radiolysis (i.e. the interaction of radiation and air and water) will change the composition of the postclosure environment.

According to present NNWSI Project package designs and package emplacement arrangements, surface temperatures on most spent fuel waste packages and in the immediately surrounding rock will remain above 96°C, the unconfined boiling point of water at this elevation, in excess of 300 years, while surface temperatures on packages containing glass waste forms and in the immediately surrounding rock may cool to 96°C some 100-200 years after emplacement. Therefore, the majority of containers will be exposed to a water vapor and air environment during a significant fraction of the containment period. Immersion of the containers is an unanticipated event in the proposed repository location during the postclosure period.

Identification of Candidate Materials

At the end of FY 1984, the NNWSI Project developed a test plan for evaluating copper and copper-base alloys as alternative container materials for a high-level nuclear waste package for disposal at a potential repository site in tuff rock at Yucca Mountain, Nevada. The candidate materials are: OFHC copper; aluminum bronze; and Cupronickel 70-30. Alloyed copper was expected to resist uniform corrosion better than copper in the oxidizing environment of a tuff repository; however, the NNWSI Project included pure copper to provide a link to the Swedish program and to serve as a baseline for comparing all the copper alloys.

NNWSI PROJECT FEASIBILITY CRITERION 1: CONTAINER DESIGN AND DEVELOPMENT

Container Design

Due to the absence of any significant hydrostatic or lithostatic pressure on the container, the NNWSI Project container thickness is based more on concerns about corrosion resistance than structural integrity. The highest expected dynamic loads on the reference container would occur during handling and retrieval. The lower strength of OFHC copper would likely require a minimum thickness of 3 cm, based on results of preliminary structural analyses; however, based on the mechanical properties of the copper alloys, a thickness of 1 cm is adequate for the aluminum bronze and Cupronickel 70-30 designs under consideration.

Acquisition of Mechanical Property Data

The NNWSI Project contracted CDA to collect and assess all relevant mechanical property data in the literature for the copper and copper-base materials of interest. This information was shared with BWIP. The data collected included short-time tensile and yield data, but most of the data were for wrought rather than cast materials. Companion data on creep properties were similarly limited. The data for pure copper were usually limited to below 250°C, whereas data for the copper alloys extend to higher temperatures. Stress rupture data were available only for the alloys. The NNWSI Project concluded that more work needs to be done to obtain the necessary mechanical property data.

Assessment of Currently Available Fabrication and Closure Methods

The Copper Development Association surveyed a number of U.S. manufacturing firms regarding current equipment and processes. The results of this survey were that the thin-walled reference design can be made relatively simply by utilizing currently available technology. The fabrication and assembly of suitable disposal containers does not appear to be a limiting factor in the use of copper-base materials.

Radiation Shielding Analysis

For 10-year old spent fuel with a burnup of 33,000 Mwd/MTU, the dose rate was calculated to be 4.8×10^4 rad/h at the container surface for a 1 cm thick container. The dose rate was reduced to 1.3×10^4 rad/h for a 3 cm thick wall. For HLW, the dose rate (10y out of reactor) at the container surface was 2.2×10^3 rad/h, dropping off to 6.3×10^2 rad/h for a 3 cm thick container.

Container Design and Development Feasibility Evaluation (Summary)

Based on the various studies and investigations performed during FY 1985 and FY 1986, the NNWSI Project believes that copper-base materials for usage as a container material pose no special challenge beyond current design and manufacturing technology.

NNWSI PROJECT FEASIBILITY CRITERION 2: PRE-CLOSURE SAFETY

Pre-closure safety refers to accident probabilities and consequences during container fabrication and emplacement. In terms of these issues, no additional hazards are introduced by the use of copper or copper alloys in a properly-designed container.

NNWSI PROJECT FEASIBILITY CRITERION 3: REPOSITORY INTERFACES (HANDLING)

The effect of the usage of copper-based containers on handling in the surface facilities or during emplacement was evaluated. It is not considered to be a limiting factor to feasibility.

NNWSI PROJECT FEASIBILITY CRITERION 4: RETRIEVABILITY

To meet the NRC requirement for retrievability, the waste container, pintle assembly, pintle attachment welds, and waste package emplacement configuration are designed to be compatible with the initial emplacement operations and any subsequent retrieval operations. Retrievability is not considered to be a limiting factor to use of a copper container.

NNWSI PROJECT FEASIBILITY CRITERION 5: EVALUATION OF CONTAINMENT POTENTIAL

Types of Copper Degradation for Consideration in the Expected Tuff Repository Environments

The types of corrosion that may develop in the tuff repository are: uniform corrosion, pitting corrosion, crevice corrosion, intergranular corrosion, selective leaching, and EAC, i.e. stress corrosion cracking and hydrogen embrittlement. Corrosion testing in irradiated environments was emphasized. Uniform and pitting corrosion were studied experimentally because they were considered the most likely degradation modes. Susceptibility to other forms of corrosion was evaluated by use of available literature data.

Uniform Corrosion

Although the copper and copper-base alloys can undergo a number of corrosion degradation mechanisms in various environments, the predominant mode of degradation expected in the tuff environment is low-temperature oxidation. Uniform corrosion in aerated wet environments may be enhanced in the presence of gamma radiation due to the production of the oxidizing species, hydrogen peroxide and nitric or nitrous acids. However, mitigating circumstances may intervene to remove some or all of oxidizing species as they are formed. For example, copper itself will catalyze the degradation of some of the hydrogen peroxide. Also, the ionic content of the groundwater is expected to buffer the nitric or nitrous acids.

Experiments were run to evaluate the effect of radiation (3.3 Mrad/h) on the uniform corrosion rate of the copper-base materials in steam and groundwater. After about 8 months exposure time, pure copper and aluminum bronze appeared to corrode at about the same rate as they did without radiation. However, the rate of corrosion of Cupronickel 70-30 appears to be approximately 10 times higher in steam in the presence of gamma radiation, with a maximum value reported at about 6 microns per yr. If the highest short-term uniform corrosion rate is linearly extrapolated to 1000 years, an extremely conservative way of projecting uniform corrosion loss, a corrosion allowance of 0.6 cm is obtained. This projection indicates that, with proper design, the general corrosion resistance of any of the copper materials would be adequate to meet the 300 to 1000 year containment requirement, if uniform corrosion were the only mechanism of concern.

Pitting Corrosion

It is possible that alterations in the environment due to gamma irradiation may produce species that promote pitting or crevice attack on copper-base materials. Tests performed in gamma irradiation in steam and normal groundwater showed no evidence of surface corrosion for pure copper but did show some indication of shallow pitting in aluminum bronze and Cupronickel 70-30. The pits appear to be superficial; however, more detailed evaluation is required to assess the depth of penetration. At this point, the observation of shallow pitting is not considered to be a disqualifying factor in the potential use of copper alloys in containers.

Containment Feasibility Evaluation (Summary)

The NNWSI Project has concentrated on studying the radiation-enhanced uniform and pitting corrosion of the three copper-based materials. Use of aqueous environments and high radiation doses have led to some enhancement of the uniform corrosion rate under these worst case conditions. Some shallow pitting has been observed in the two alloys studied in the same environment. However, based on the limited test results to date of the NNWSI Project two-year feasibility assessment program and an evaluation of the literature that exists on the corrosion of copper materials in non-radiation environments, it has been concluded that the corrosion degradation observed in laboratory tests and the corrosion behavior postulated on the basis of current knowledge are not sufficiently serious to cause copper-based materials to be excluded from usage as a container material in the tuff repository.

NNWSI PROJECT FEASIBILITY CRITERION 6: EVALUATION OF EFFECTS OF COPPER ON RADIONUCLIDE RELEASE CHARACTERISTICS

A further consideration regarding the feasibility of copper is whether it can increase the release of radionuclides to the environment. One factor affecting this release would be a detrimental interaction between copper and Zircaloy spent fuel cladding when the containment period is over, the container has been breached, and an aqueous environment is present. In order to assess this potential effect, Zircaloy cladding samples were placed in contact with a wrap of copper foil material. The assembly was set in a bed of crushed tuff in a container filled with copper nitrate or groundwater. After 5 months exposure, the cladding was not degraded. Thus, results to date do not indicate that copper would cause galvanic corrosion of the Zircaloy cladding. Longer-term tests are required as well as tests with the fuel matrix present to determine any detrimental effects of copper on release of radionuclides.

NNWSI PROJECT FEASIBILITY CRITERION 7: COST AND AVAILABILITY OF COPPER-BASED MATERIALS

The results of the CDA studies indicate that the present cost of a full-size rolled and welded prototype the NNWSI Project container would range from $28,000 to $33,000. The thicker oxygen-free copper version would have the highest cost; costs for the copper-base alloys would be the same. Estimated savings based on production volumes would amount to approximately 25%, due mainly to volume purchase of starting materials. Assessments of cost significance are difficult without employing relative measurements or comparisons. However the container cost is not expected to impact copper container feasibility for a tuff repository.

SUMMARY OF COPPER CONTAINER FEASIBILITY EVALUATION (NNWSI PROJECT)

It is concluded that copper is feasible for use as a waste container for a repository in tuff, based on the limited investigations conducted to date. However, longer-term experiments are needed to more definitively assess the feasibility of using copper-base materials in containers, particularly in regard to the effect of gamma radiation on the uniform corrosion rate. The mechanical properties of the copper-base materials, as known, do not appear to limit the feasibility of the use of these materials in containers. Fabrication, welding and inspection surveys indicate that most manufacturers feel that copper-base containers are within current manufacturing capabilities. Results to date do not indicate that copper would cause galvanic corrosion of the Zircaloy cladding. Further effects on release of radionuclides due to copper will have to be assessed. There are no apparent concerns with handling copper-base material either in the surface facilities or during emplacement, nor is there any apparent impact on retrievability or pre-closure safety.

PLANS FOR FURTHER EVALUATION OF THE USE OF A COPPER-BASE CONTAINER IN A TUFF REPOSITORY

The current list of candidate materials for a container in a tuff repository is composed of three stainless steels and three copper-base materials as follows: AISI 304L; AISI 316L; Alloy 825; OFHC copper; aluminum bronze; and Cupronickel 70-30. Based on the results of the copper feasibility study, the NNWSI Project will continue metals testing on these six materials until one material is selected based on weighted selection criteria developed by the NNWSI Project.

The copper feasibility study has revealed a number of areas where further testing and evaluation are warranted. These areas include, for example, extended Zircaloy interaction studies, copper/waste form interaction studies, plus longer-term uniform and localized corrosion studies, particularly in moisture films and under gamma irradiation; microbiological and embrittlement effects, if any, on corrosion; and comprehensive fabrication, closure and inspection studies. All of these additional studies will be assessed in FY 1988, and tests being conducted on the final candidate material(s) may be extended on a long-term basis.

SUMMARY

A two-year feasibility assessment of the use of copper-base materials in high-level waste containers in basalt and tuff repository environments has been completed. The evaluation criteria in the assessment were design and development, pre-closure safety, repository interfaces, retrievability, potential for long-term containment, effect on radionuclide release, and cost and availability. The BWIP and the NNWSI Project concluded that copper-base materials are feasible as candidate container materials in a repository sited in a basalt or tuff environment. The BWIP and the NNWSI Project will continue their evaluations of the copper-based materials until a decision on choice of container material is made.

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