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Civilian Radioactive Waste Management System
Management & Operating Contractor

Waste Package Neutron Absorber, Thermal Shunt, and Fill Gas Selection Report

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Revision History

Revision number	Date	Description of the change
00	12-10-99	Initial issue. It supercedes Waste Package Materials Selection Analysis BBA000000-01717-0200-00020 REV 01, December 1997.

EXECUTIVE SUMMARY

Materials for neutron absorber, thermal shunt, and fill gas for use in the waste package were selected using a qualitative approach. For each component, selection criteria were identified; candidate materials were selected; and candidates were evaluated against these criteria. The neutron absorber materials evaluated were essentially boron-containing stainless steels. Two candidates were evaluated for the thermal shunt material. The fill gas candidates were common gases such as helium, argon, nitrogen, carbon dioxide, and dry air. Based on the performance of each candidate against the criteria, the following selections were made:

Neutron absorber	Neutronit A978
Thermal shunt	Aluminum 6061 or 6063
Fill gas	Helium

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1. INTRODUCTION

1.1 BACKGROUND

According to the Uncanistered Spent Nuclear Fuel Disposal Container System Description Document (comes from SDD-UDC-SE-000001 REV 00, Volume I, sections 1.2.2.1.12 and 1.2.1.6, at p. 13 and p. 11 respectively), the waste package (WP) must be designed such that, under normal condition(s), nuclear criticality does not occur and temperature of the Zircaloy cladding remains below 350°C. To fulfill these requirements, the WP must contain inert atmosphere within it, and it must also be designed to possess in-built system(s) to reduce neutron concentration and support rapid transfer of heat towards outside. Accordingly, the waste package design selected for the Viability Assessment (VA) included a neutron absorber material, thermal shunt, and fill gas as part of the internal constituents. Material selections for these elements are being reevaluated in view of the changes in waste package design for the Site Recommendation (SR) and License Application (LA). Considering the expected long life of the waste package, particular emphasis has been given to the selection of materials with high corrosion resistance for application as neutron absorber and thermal shunt. This report is one of a series of reports prepared on the waste package materials selection in support of the SR/LA effort.

1.2 OBJECTIVE

The primary objective of this study is to evaluate candidates for the neutron absorber, thermal shunt, and fill gas for the waste package, leading to the selection of the most suitable material for each application.

1.3 SCOPE

The scope of this project will include identification and evaluation of candidate materials for the neutron absorber, thermal shunt, and fill gas for use as part of the internals of the waste package. The evaluation procedure will include identification of the functional requirements of each component, selection criteria, evaluation of each candidate material, and selection of the most suitable material for each component.

1.4 QUALITY ASSURANCE

This technical document was prepared in accordance with QAP-3-5, *Development of Technical Documents* (CRWMS M&O 1999d). The information provided in the technical document is to be indirectly used in the evaluation of the Mined Geologic Repository (MGR) waste package and engineered barrier segment. The waste package and engineered barrier segment have been identified as Quality Level 1 items in the QAP-2-3 evaluations (e.g., *Classification of the MGR Uncanistered Spent Nuclear Fuel Disposable Container System*) (CRWMS M&O 1999a). The Waste Package Materials Department responsible manager has evaluated the technical document development activity in accordance with QAP-2-0, *Conduct of Activities*. The QAP-2-0 activity evaluation, *Activity Evaluation SR Report Vol 1* (CRWMS M&O 1999b,) has determined that the preparation and review of this technical document is subject to *Quality Assurance Requirements and Description* (DOE 1998) requirements. There is no determination of importance evaluation developed in accordance with Nevada Line Procedure, NLP-2-0, *Determination of Importance Evaluations*, since the report does not involve any field activity.

1.5 USE OF COMPUTER SOFTWARE

No computer software was used.

2. DESCRIPTION OF THE WASTE PACKAGE NEUTRON ABSORBER, THERMAL SHUNT, AND FILL GAS

The most important function of the neutron absorber is to absorb neutrons and reduce the potential for criticality. The neutron absorber material is typically an additive material to a carrier material (e.g., stainless steel alloyed with a boron compound). The neutron absorber (carrier material) is used as part of the internal component structure and is designed as interlocking plates within the spent-fuel basket structure. Thermal shunts may be needed in some of the waste packages that may contain fuel assemblies with relatively high decay heat. The thermal shunt, when used, provides an important additional high thermal conductivity path for conducting heat from the fuel to the containment barriers. Thus, the thermal shunt performs an important function in maintaining the spent fuel material below the designated temperature. The fill gas function is to provide an inert atmosphere within the waste package and facilitate heat conduction from the waste form to the containment barriers.

3. DESCRIPTION OF THE INTERNAL COMPONENTS MATERIAL SELECTION PROCESS

3.1 SELECTION OF NEUTRON ABSORBER MATERIAL

3.1.1 Selection Criteria

The functions of this component were discussed in Section 2. Along with its most important functional requirement of providing criticality control, this component must be able to sustain mechanical loads due to handling, emplacement, and, if necessary, retrieval, but these loads are not especially large. Therefore, mechanical performance is selected as one of the criteria. Corrosion behavior is important in keeping the neutron absorber material in place and effective in controlling criticality long after emplacement. So chemical performance in a variety of environments will be used as an important selection criterion. The material should not degrade other components, but its function of criticality control will not be significant unless a moderator (water) is present in the waste package, and, for that to happen, the containment barriers must already have failed. Therefore, compatibility only requires that the basket plates not degrade the waste form. The fuel basket plates provide a substantial fraction of the cost of the disposal container, so a moderate importance is assigned to cost. The fuel basket plates also provide an important path for conducting heat from the fuel to the containment barriers; therefore, thermal performance is also a criterion.

This evaluation is intended to be a qualitative assessment of each candidate material against the noted criteria; therefore, no numerical weighting factors are being assigned to the criteria. Instead, the evaluation will include positive and negative attributes of each candidate (as related to the criteria) based on engineering judgement.

3.1.2 Neutron Absorber Candidate Materials

In view of the corrosion resistance required of the neutron absorber materials, the candidate materials selected for evaluation are primarily stainless steels of various types. These included ASTM (American Society for Testing and Materials) A 887 Type 304B3 Grade A, ASTM A 887 Type 304B4 Grade A, ASTM A 887 Type 304B5 Grade A, ASTM A 887 Type 304B6 Grade A, Neutronit A976 with 1.6% boron, and Neutronit A978 with 1.6% boron. Neutronit A976 and A978 are grades of plate produced by Böhler Bleche GmbH of Mürzzuschlag, Austria. The various types of ASTM A 887 Grade A are apparently produced only by Carpenter Technology Corporation of Reading, PA.

A variety of additional materials were considered in a recent corrosion test (Van Konynenburg and Curtis 1995). These materials (Konynenburg and Curtis 1995, Table 2) may be classified as follows: aluminum-matrix composites, copper-matrix composites, austenitic stainless steel without boron, austenitic stainless steels with boron, zirconium-hafnium alloys, and nonmetallic materials (ceramics or minerals). Of these materials, only the austenitic stainless steels with boron are acceptable for fabrication of fuel basket plates. Aluminum-matrix composites and copper-matrix composites have unacceptably high corrosion rates (Konynenburg and Curtis 1995, Abstract). In particular, BORAL is subject to hydrogen gas production, deformation, and delamination (Smith et al. 1992, p. 3-1). Austenitic stainless steel without boron was a control material and was not intended for fabrication of basket plates. Zirconium-hafnium alloys are unacceptable because of the high cost and limited availability of hafnium. The nonmetallic materials are brittle and would not provide acceptable mechanical performance.

3.1.3 Evaluation of the Candidate Materials

Mechanical performance–Mechanical performance is defined by the combined effects of material strength and ductility. "Strength" is the minimum yield strength as given in appropriate ASTM specifications. "Ductility" is the minimum elongation as given in the specification. In general, higher strength and higher ductility are preferable. The yield strength provides a measure of the loads that the material can withstand without significant permanent distortion. Yield strength and ductility together provide a conservative estimate of the energy that the material can withstand without failing. The mechanical properties of the candidate materials are shown in Table 3-1.

Table 3-1. Mechanical Properties of the Candidate Materials

Material	Yield Strength (MPa)	Elongation (%)	Reference
ASTM A 887 Type 304B3 Grade A	207	31	ASTM 1990, Table 2
ASTM A 887 Type 304B4 Grade A	207	27	ASTM 1990, Table 2
ASTM A 887 Type 304B5 Grade A	207	24	ASTM 1990, Table 2
ASTM A 887 Type 304B6 Grade A	207	20	ASTM 1990, Table 2
Neutronit A976	300	9	CRWMS 1999c, pp. 39 and 40
Neutronit A978	300	9	CRWMS 1999c, pp. 39 and 40

A review of the above data suggests that all of the candidate materials have adequate strength for the expected service loads.

Chemical performance–For conditions that are relevant to repository conditions, less information on corrosion of the candidate materials is available for this component than for the others. The following information on corrosion performance is noted: Van Konynenburg and Curtis (1995) exposed samples of Böhler A976 SD (Neutronit A976), Neutrosorb Plus (ASTM A 887 Grade A, type not specified) base metal, and Neutrosorb Plus (ASTM A 887 Grade A, type not specified) welded metal to an aqueous mixture of formic acid, sodium formate, sodium oxalate, nitric acid, sodium chloride, and hydrogen peroxide in short term corrosion tests. They observed corrosion rates of 41 μm/yr for corrosion of the Neutronit A976, 60 μm/yr for the ASTM A 887 Grade A base metal, and 880 μm/yr, with pitting, for the ASTM A 887 Grade A welded metal (Konynenburg and Curtis 1995, Table 3). Smith et al. (1992) also reports on several corrosion tests. These include corrosion rate in 65% nitric acid (Smith et al. 1992, Table 3-15 and Table 3-19), intergranular corrosion tests in 6% copper sulfate plus 16% sulfuric acid (Smith et al. 1992, Table 3-16), tests in 2000 ppm H_3BO_3 (Smith et al. 1992, Table 3-17), 5% sodium chloride spray, and 5% ferric chloride (Smith et al. 1992, Table 3-18), and ferric sulfate plus sulfuric acid (Smith et al. 1992, Table 3-19). However, the results reported in Smith et al. (1992) were not intended to address repository conditions.

A significant difference between the candidate materials for this component as compared to those for other components is that the compositions of candidate materials for this component are much more tightly clustered. Because of this and because limited corrosion data are available, chemical performance is, therefore, evaluated by invoking analogous materials of similar composition but without boron.

For the ASTM alloys, compositions can be compared between ASTM (1990, Table 1) and CRWMS (1999c, p. 17). The composition limits for carbon, manganese, phosphorus, sulfur, silicon, chromium, and nitrogen are identical for ASTM A 887 Types 304B3 through 304B6 and ASTM A 240 Type 304. The range of allowable nickel content is higher for ASTM A 887 Types 304B3 through 304B6 than for ASTM A 240 Type 304. This is taken as evidence that the composition without boron of ASTM A 887 Types 304B3 through 304B6 is based on that of ASTM A 240 Type 304.

For the Neutronit alloys, the manufacturer's information provides average values for some elements and maxima for others. For Neutronit A976, the specified values for carbon, chromium, and nickel are all within the specification limits of ASTM (1990, Table 1). The limit on carbon is more stringent than that of ASTM A 887, and a limit on cobalt is added. This is taken as evidence that the composition without boron of Neutronit A976 is similar to that of the alloys specified in ASTM A 887. Those alloys in turn have already been shown to be based on ASTM A 240 Type 304, so the composition without boron of Neutronit A976 is based on that of ASTM A 240 Type 304.

For Neutronit A978, the specified values for carbon, nickel, and molybdenum are all within the specification limits of CRWMS (1999c, p. 17) for ASTM A 240 Type 316. The limit on carbon is more stringent than that of CRWMS (1999c, p. 17), and a limit on cobalt is added. The average chromium content is slightly above the range specified in CRWMS (1999c, p. 17). This is taken as evidence that the composition without boron of Neutronit A978 is similar to that of ASTM A 240 Type 316.

The above evaluation suggests that Neutronit A978 will exhibit better corrosion resistance than all of the other candidate materials because of its similarity to Type 316 stainless steel which is more corrosion resistant than Type 304.

Neutronic performance–For all of the candidate materials, boron is the element that contributes most prominently to neutron absorption. Accordingly, the neutronic performance is rated by the amount of boron in the alloy. Of the candidate materials evaluated, the Neutronit alloys have the highest amount of boron (1.6%).

Compatibility with other materials–The function of preventing criticality is not significant unless the containment barriers fail and large amounts of liquid water enter the degraded waste package. Therefore, before the fuel basket plates begin to provide this function, the remaining components will be severely degraded and will not provide significant functionality. As a result, this component need only be compatible with the waste form. Stainless steels are used in proximity with nuclear fuel assemblies. This is taken as evidence that the stainless steels will not degrade the waste form. Therefore, it is judged that all of the candidate materials are compatible with the waste form.

Thermal performance–Evaluations of thermal performance were based on room temperature thermal conductivities. Thermal conductivities for these materials are difficult to find. CRWMS (1999c, p. 39) gives a thermal conductivity of 10.3 W/m·K at 20 °C for the Neutronit alloys. No distinction is made between the two alloys, so their thermal conductivities are presumably similar.

Thermal conductivities were not found for the ASTM alloys. Based on the composition of the matrix, these may have a slightly higher thermal conductivity than Neutronit A978. As is discussed above, the ASTM A 887 alloys and Neutronit A976 have compositions that are based on ASTM A 240 Type 304, whereas Neutronit A978 has a composition that is based on ASTM A 240 Type 316. For comparison, CRWMS (1999c, p. 18) gives thermal conductivities of 14.88 W/m·K for Type 304 and CRWMS (1999c, p. 15) gives a value of 13.33 W/m·K for Type 316, both at about 300 K. The addition of borides appears to reduce the thermal conductivity. This information on thermal conductivity is taken as evidence that all six candidate materials have similar thermal conductivities

Cost–The cost of all of the candidate materials are very close to each other, and, thus, the cost is not a meaningful discriminator for the selection.

In a review of the information presented above, it appears that the most important attributes of the performance of the candidate materials are corrosion resistance and available boron content. Based on this, it is recommended that Neutronit A978 be selected as the material for the neutron absorber plates at this time.

In the recent past, several additional materials have been suggested. These include gadolinium oxide and/or gadolinium phosphate, thermally sprayed on a metal substrate such as stainless steel, and zirconium clad boron carbide rods, etc. At this time these are in the conceptual stage. It is recommended that additional data be obtained on these concepts to establish their viability.

3.2 SELECTION OF THERMAL SHUNTS

With the expected changes to the repository and waste package designs for SR/LA, it is not clear if thermal shunts will be needed to meet the temperature limits on the spent fuel cladding. However, this section addresses selection of materials for this component, should there be a need for the use of thermal shunts.

3.2.1 Selection Criteria

As mentioned earlier, the primary function of the thermal shunt is to conduct heat from the waste form to the containment barrier. Therefore, thermal conductivity of the materials is very important. These components are needed during the early periods of the repository storage (few hundred years) when the decay heat of the spent fuel is relatively high. Thus, the material selected need not have a high degree of corrosion resistance. The thermal shunts are required to have adequate structural strength to withstand handling, emplacement, and possible retrieval operations. However, these service loads are not very large, so the mechanical performance was not selected as an evaluation criterion.

The thermal shunts, by the nature of their function, are in contact with the waste form. Therefore, compatibility with the spent fuel assemblies was the other criterion selected for evaluation.

3.2.2 Candidate Materials

In view of the very limited functional requirements on the thermal shunts, only two candidate materials were evaluated. These are aluminum and aluminum alloys and copper. Both classes of these materials have excellent thermal conductivities, and both are relatively inexpensive.

3.2.3 Evaluation of the Candidate Materials

Thermal performance–Evaluations of thermal performance were based on room temperature thermal conductivities. Aluminum alloys 6061 and 6063 are nearly all-aluminum with very similar thermal properties. The thermal conductivity of these materials range from about 166 to 209 W/m·K (CRWMS 1999c, pp. 56 and 61). The thermal conductivity of pure copper is given as 339.2 W/m·K (CRWMS 1999c, p. 68). Based solely on thermal conductivity, copper would be a better choice for the thermal shunt.

Compatibility with waste form–Compatibility of the thermal shunt with the waste form is of concern when the waste package containment barrier is breached, and water enters the waste package. Under these circumstances, the thermal shunt should not degrade the waste form. Aluminum alloys are expected to corrode sacrificially when coupled with highly corrosion resistant zirconium alloys. Copper, on the other hand, may react with the chloride ions in the water and produce cupric chloride which is known to cause localized corrosion in zirconium alloys. This could potentially result in an accelerated degradation of the cladding and consequential release of radionuclides from the fuel rods. This suggests that copper will not be suitable for use as a material of choice for the thermal shunt.

3.3 SELECTION OF FILL GAS WASTE PACKAGE

3.3.1 Selection Criteria

The fill gas can be a significant conductor of heat from the fuel to the basket, and therefore, thermal performance is deemed one of the most important criterion. The fill gas should not degrade other components of the waste package, so compatibility with other materials is another important criterion. Controlling cost is desirable, but fill gas is believed to be an inexpensive part of the waste package.

3.3.2 Candidate Materials

Candidate materials include a vacuum plus a variety of common gases: helium, carbon dioxide, nitrogen, oxygen, argon, dry air, and environmental air. Classification of vacuum as a gas is arbitrary and serves only to provide a comparison. To avoid questions of compromising the underground facility, the fill gas should not be explosive or chemically reactive. This constraint clearly excludes oxygen (although air contains some oxygen).

3.3.3 Evaluation of the Candidate Materials

Thermal performance–The criterion for thermal performance of the fill gas is its thermal conductivity (the various gases are compared at 600 K and 101 kPa). Thermal conductivity of the gas is most important at high temperature because of the thermal goal of limiting fuel cladding temperature. The following table provides the conductivity data for the selected gases.

Table 3-2. Thermal Conductivities of Gases at 600 K and 101 kPa

Material	Thermal conductivity (W/m·K)	Reference
Helium	0.2524	CRC (1995, p. 6-251)
Carbon dioxide	0.0416	CRC (1995, p. 6-251)
Nitrogen	0.0440	CRC (1995, p. 6-251)
Argon	0.0306	CRC (1995, p. 6-251)
Dry air	0.0457	CRC (1995, p. 6-251)

The above data shows that helium is the most suitable choice for the fill gas from the thermal performance standpoint.

Compatibility with other materials–Vacuum is chemically inert. Helium and argon are noble gases, and noble gases are essentially chemically inert, so they would cause negligible damage to the waste package. Helium is routinely used as the fill gas for the fuel rods, indicating it has excellent compatibility with the spent fuel. Radiolysis of carbon dioxide might conceivably liberate oxygen which could oxidize fuel with failed cladding. For waste packages filled with nitrogen, radiolysis of nitrogen and water from waterlogged fuel might produce sufficient quantities of nitric acid for condensation of corrodents to take place. Carbon dioxide and nitrogen may cause damage if radiolysis is significant. Because of their nitrogen content, dry air and environmental air have the same disadvantages as nitrogen, plus the disadvantage that oxygen in the air could oxidize fuel with failed cladding. Dry air and environmental air can cause damage under certain conditions.

Based on a review of the data on thermal conductivity and material compatibility, the most suitable material of choice for the fill gas is helium.

4. CONCLUSIONS

Based on the evaluations presented in Sections 3.1, 3.2, and 3.3, the following conclusions are obtained.

Material of choice for the neutron absorber is Neutronit A978. This selection is based on the corrosion performance of this material compared to the other candidate materials and available boron concentration.

Aluminum alloy 6061 or 6063 was selected as the material for thermal shunt. The other candidate material copper was rejected for possible compatibility problems with the spent fuel cladding.

Helium was selected as the material for waste package fill gas. Helium has been in use as the fill gas for fuel rods and has been found to be highly compatible with the fuel material. It is also an excellent conductor of heat.

5. REFERENCES

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