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DPC Materials and Corrosion Environments

Fuel Cycle Research & Development

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ACRONYMS

AISI	American Iron and Steel Institute
BSC	Bechtel-SAIC Co.
BWR	Boiling Water Reactor
CS	Carbon steel
DOE	U.S. Department of Energy
DPC	Dual Purpose Canister
DU	Depleted uranium
HAZ	Heat-affected zones
ISO	International Standards Organization
MMC	Metal matrix composite
NRC	Nuclear Regulatory Commission
MPC	Multi-Purpose Canister
PWR	Pressurized Water Reactor
SCC	Stress Corrosion Cracking
SNF	Spent Nuclear Fuel
SS	Stainless steel
UNF	Used Nuclear Fuel
UFD	Used Fuel Disposition

DPC MATERIALS AND CORROSION ENVIRONMENTS

1. Materials Used in DPCs

Canister shells are nearly all constructed from stainless steel alloys, with the exception of some versions of the NAC-MPC canister that use carbon steel (Table 1). Typical canister shells are made from stainless steel SS-304L or 316 (Hardin et al. 2013). Holtec International lists “alloy X” as the preferred canister material, where the four stainless steel alloys that meet the requirements for “alloy X” are 316, 316LN, 304, and 304L (BSC 2003).

Baskets maintain spacings between fuel assemblies, transfer heat, and support neutron absorber elements. A number of common DPC designs use stainless steel for major basket components (e.g., Holtec HI-STAR 100 MPC®, NAC Universal MPC® system, and most of the NUHOMS® systems) (BSC 2003; Greene et al. 2013). In general, baskets are fabricated with square stainless steel tubes, or egg-crate arrangements of stainless steel plates, to which sheets of neutron absorbing material are affixed between assemblies. Besides stainless steels 304/304L and 316/316L, other basket materials can include: precipitation-hardened Type 17-4 (for PWR support disks), SA-533 (for BWR support disks), and aluminum (for heat transfer disks) (NAC UMS and Yankee systems; BSC 2003). In some NUHOMS® systems, aluminum-coated carbon steel is used to support flux traps in the basket, and in the BNFL Fuel Solutions DPC some of the basket components can be fabricated with nickel-coated carbon steel (BSC 2003).

Metal matrix composite (MMC) materials are commonly used for neutron absorbers. MMC sheets are fabricated with finely divided boron carbide (B4C) evenly dispersed in an aluminum matrix (Lindquist 2009). Commonly used MMC materials (e.g., Boral® or Metamic®) perform in fuel pools and are used in DPCs licensed for transportation, but can readily corrode over time frames relevant to disposal: on the order of 1.91 mg cm-2 yr-1 giving estimated corrosion lifetime of as few as 40 years (Lindquist 2009). Accordingly, these materials cannot be relied on as neutron absorbers during long-term exposure to ground water in geologic disposal environments.

More durable neutron absorber materials include borated stainless steel and nickel-gadolinium alloys (Wells 2008). Laboratory testing using electrochemical methods show estimated general corrosion rates for these materials ranging from 0.03 to 0.65 µm yr-1 for borated stainless steel 304B4, and from 0.96 to 94 µm yr-1 for a Ni-Cr-Mo-Gd alloy (Lister et al. 2007). A few DPC systems offered by vendors would use longer-lasting neutron absorber materials, but implementation has been very limited and is not significant with respect to the overall inventory of existing DPCs (BSC 2003; Greene et al. 2013).

Whereas aluminum-based materials such as Boral® or Metamic® are expected to readily corrode in any disposal environment where exposed to ground water, stainless steel basket structures could maintain configuration stability depending on corrosion modes and rates, and basket component thicknesses. Structural integrity is unlikely to be significantly affected by localized corrosion (crevice corrosion, pitting, and stress corrosion cracking) especially for basket components that are loaded in compression. The basket fits closely into the canister shell, which provides confinement, and the shell fits into the disposal overpack, which provides additional confinement.

Details of basket degradation are beyond the scope of the current study, and are called out as information needs (Sections 10.4 and 10.5). This review focuses on the performance of basket materials that could be exposed to ground water over thousands of years, and prospective disposal overpack materials that could possibly be used to protect DPCs in disposal environments.

Table 1. DPC materials summary (data from Greene et al. 2013)

Storage/Transport System	Canister Materials	Internals/Basket Materials	Shield Plug Materials
FuelSolutions™	SS	Steel/Boral®	DU/SS or SS/CS
HI-STORM 100®	SS	SS/Metamic®/Al	SS
HI-STAR 100®	SS	SS/Boral®/Al	SS
		SS/Metamic®/Al	SS
HI-STAR 190®	N/A	N/A	
NAC-MPC®	SS	SS/Boral®/Al	SS
	Concrete/CS	NS-4-FR	SS
	CS/Pb	NS-4-FR	SS
NAC-UMS®	SS	SS/Al/Boral®	SS
	SS	SS/CS/Boral®/Al	SS
	Concrete/Steel	NS-4-FR	NS-4-FR
NAC-MAGNASTOR®	SS	Ni-plated CS/SS	SS
NUHOMS®	SS	Steel	CS or SS/Pb
	SS	Steel/B-Al/Al	Steel/Pb
	SS	SS/Boral®	Steel or Steel/Pb
	SS	SS/B-Al/Boral®/MMC	Steel
	SS	SS/Al/MMC	Steel
	SS	SS/Al-B sheets	CS or Steel/Pb
	SS	SS/Boron plates	CS
	SS	SS/Boral®	SS/Pb
	SS	CS/B-SS	CS
	SS	CS/B-Al/MMC/Boral®/Al	CS

Note: CS = carbon steel; SS = stainless steel; DU = depleted U; other symbols are trademarks as noted, or chemical elements.

2. Factors Controlling Material Corrosion Rates in Disposal Environments

Based on the driving forces and mechanisms, the following corrosion types have been identified: galvanic or bimetallic, uniform or general attack, localized corrosion (crevice, pitting, intergranular, and stress corrosion cracking; APV 2008), and microbial. The local geochemical environment plays a significant role in corrosion processes, and prominent physico-chemical drivers include availability of moisture, ion concentrations, pH, and temperature.

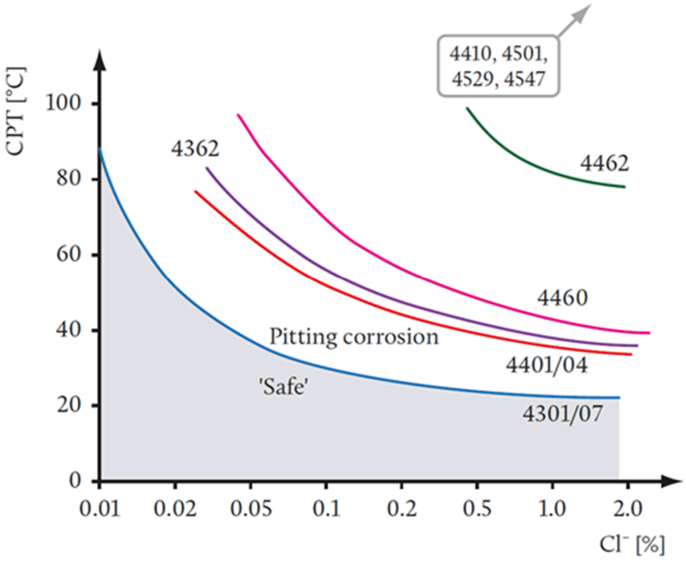
Galvanic corrosion occurs when two conducting materials are in electrical contact with each other in an electrolyte solution. As a result, galvanic current is generated between these two metals. This galvanic current generally causes a reduction in the total corrosion rate of the cathodic member of the couple, while amplifying the corrosion of the anode (which is sometimes called the sacrificial anode) (Zhang 2011). This process is well understood for various metallic material couples, and is not expected to impact stainless steel. The aluminum-based neutron absorbers will served as protective anodes for stainless steel basket materials (one reason why these neutron absorbers would be relatively short-lived). For stainless steel to be anodically corroded would require a cathode that is significantly more noble (higher on the galvanic series).

Corrosion resistant materials such as copper, titanium, Zircaloy and nickel-chromium alloys that might be used in the disposal overpack, do not produce sufficient potential differences with stainless steel to support galvanic corrosion. Hence, galvanic corrosion would not be important to long-term behavior of stainless steel in DPCs, in disposal environments.

Uniform or general corrosion is characterized by both anode and cathode reactions across the entire surface of the material. As a result, a overall uniform loss of material (due to the oxidation and dissolution of the oxide layer) is observed. In corrosion allowance materials such as carbon steel, uniform corrosion is the most common corrosion type yet it is considered negligible in many industrial applications. Uniform corrosion is slow in corrosion-resistant materials (generally passivating metals, including stainless steels) and is generally detectable only in aggressive chemical media such as concentrated acids or bases (Jessen 2011).

Localized corrosion can take place within days, and therefore can significantly limit containment lifetime of materials such as stainless steel (although it may be of minor importance to basket structural lifetime as discussed above). For example, pitting penetration of a 0.5 mm thick stainless sheet in the presence of chloride and hydrogen peroxide was observed after 4 days (Jessen 2011). Pitting and crevice corrosion are the most common localized corrosion types for stainless steels.

The formation of pits on stainless steel surfaces proceeds due to the local breakdown of the passive oxide layer. The pit behaves as an anode, while the oxide film over the rest of the surface behaves as a cathode, with the resulting internal galvanic coupling driving pit growth (Jessen 2011). Temperature, chloride concentration, availability of oxidants, and pH are the most important chemical drivers for local corrosion including pitting (Kursten et al. 2004; Smart et al. 2004). The corrosion rate increases with increasing temperature, chloride content, availability of oxidants (e.g., hydrogen peroxide, perchlorate, dissolved oxygen), and decreasing pH. Laboratory measurements of stainless steel pitting in well-aerated, semi-neutral solutions show that concentration of chloride controls the critical pitting temperature. Critical pitting temperature curves as a function of chloride content for several stainless steel alloys are shown in Figure 1. However, it should be noted that these data are for the base metals; sensitization and residual stresses in weld heat-affected zones (HAZ) lower the critical pitting temperature for a given chloride concentration.



Notes:

1. ISO grades shown.
2. ISO4401 is comparable to Grade SS 316, and ISO4301 is comparable to SS 304.

Other grades shown are high-performance austenitic and/or martensitic compositions.

Figure 1. Critical pitting temperature for stainless steel alloys as a function of chloride concentration (from Jassen 2011)

Crevice corrosion is controlled by the same chemical drivers as pitting. However, these two corrosion processes differ in how fluids are replaced: crevice corrosion evolves in confined spaces where the fluid is replaced by diffusion, while fluids are free to move convectively during pitting corrosion (Jassen 2011). Diffusion-limited fluid transport causes the evolution of localized conditions favoring enhanced corrosion (e.g., lower local pH and ion concentrations; APV 2008). Linear dimensions of the confined space determine whether crevice corrosion is likely to initiate; it usually occurs in gaps of a few hundred microns or less, and is rarely observed in crevices deeper than 2 mm (APV 2008). The initiation of crevice corrosion can happen within a few hours to several months, and can proceed rapidly after initiation (APV 2008).

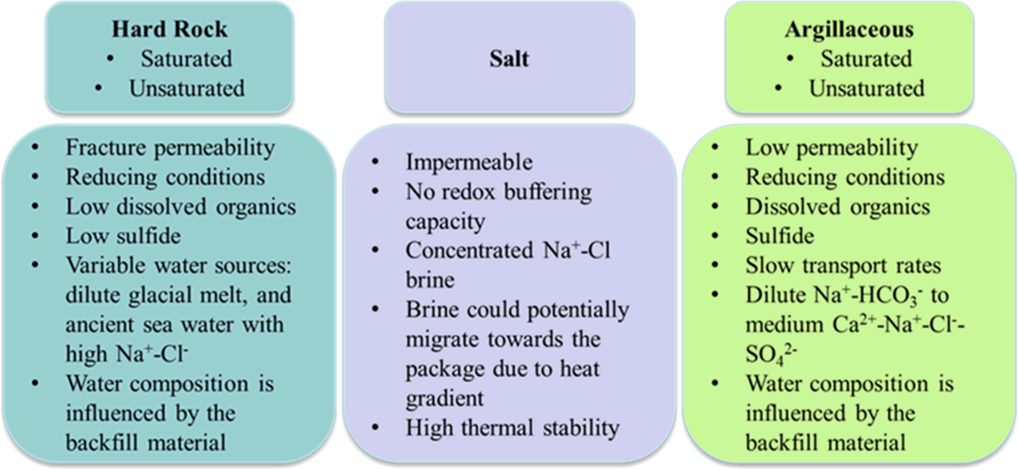
Stress corrosion cracking (SCC) is observed in corrosion resistant materials including stainless steel. It is localized around areas of residual stress produced from welding, thermal cycling, or mechanical stress to yield (cold-work). SCC can cause rapid penetration and loss of structural integrity in some applications. Similar to the other localized corrosion processes, SCC is controlled by the presence of water vapor or an aqueous phase, corrosive species (particularly chloride), oxidizers, pH, and temperature.

In addition to the five main corrosion drivers (moisture, pH, temperature, oxidizers, and chloride) a variety of geochemical constituents can significantly enhance or inhibit the rate and extent of general and localized corrosion. Metal corrosion in soils (which can be analogous to repository environments) is a multi-scale, multivariate process controlled by the local chemical environment at the corroding surface. The geometry and liquid phase chemistry of surface films/droplets and the thickness and composition of the oxide layer affect the diffusion of oxidizing species to the corroding surface (Cole and Marney 2012).

Chemical species that are not redox active, and do not attack the passive layer, can potentially decrease corrosion (Jack and Wilmott 2011). For example, the presence of dissolved organic compounds can significantly reduce corrosion rates by blocking the surface from the aggressive species (Jassen 2011). Similarly, dissolved sulfate (SO_4^{2-}) and phosphate (PO_4^{3-}) can also adsorb and protect the surface (Jassen 2011). Under normal conditions sulfate acts as a corrosion inhibitor; however, both sulfate and phosphate can serve as nutrients for microbial communities, and thereby contribute to microbially influenced corrosion (Jack and Wilmott 2011).

Microbially influenced corrosion has been described for stainless steels, and has been observed in piping in the waste water plants. Some types of bacteria can significantly lower the corrosion resistance of stainless steel. Microbial biofilms can act as crevices and change the local environment at the corroding surface (Jassen 2011). Microbial populations are ubiquitous in the subsurface, including prospective hard rock and argillaceous repository types, and possibly salt host media as well. Microbial processes therefore need to be considered when evaluating material performance and expected corrosion rates.

The following sections summarizes major physico-chemical corrosion drivers in the three repository types: hard rock (crystalline), salt, and argillaceous rock. The general geochemical features of these repository types are summarized in Figure 2. The range of groundwater compositions encountered in these repository settings is described in Table 2.



Hard Rock
- Saturated
- Unsaturated

- Fracture permeability
- Reducing conditions
- Low dissolved organics
- Low sulfide
- Variable water sources: dilute glacial melt, and ancient sea water with high Na^+-Cl^-
- Water composition is influenced by the backfill material

Salt

- Impermeable
- No redox buffering capacity
- Concentrated Na^+-Cl brine
- Brine could potentially migrate towards the package due to heat gradient
- High thermal stability

Argillaceous
- Saturated
- Unsaturated

- Low permeability
- Reducing conditions
- Dissolved organics
- Sulfide
- Slow transport rates
- Dilute Na^+-HCO_3^- to medium Ca^{2+}-Na^+-Cl^--SO_4^{2-}
- Water composition is influenced by the backfill material

Figure 2. General disposal environments and geochemical conditions

Hard Rock (Crystalline) Disposal Environments

Hard rock host media are also referred to as crystalline, or granite, and are being pursued by Swedish (Marsal et al. 2007) and Finnish (Andersson et al. 2007) repository programs. Hard rock repositories can be either saturated or unsaturated depending on the depth of the water table. They may have low or high fracture permeability, and oxidizing or reducing conditions. Concentrations of dissolved organics and sulfide are typically low. The composition of infiltrating waters is variable, but can be bounded by two chemical end members selected here: glacial melt or ancient trapped sea water.

Table 2. Range of geochemical conditions in three main geologic repository types (concentration values in mg/L, temperature in degrees Celsius)

Hard rock (granite)													
	pH	Temp.	Na^+	K^+	Ca^{2+}	Mg^{2+}	Cl^-	HCO_3^-	NO_3^-	SO_4^{2-}	HS^-	F^-	PO_4^{3-}
High	-	-	63900	17678	108747	15306	212280	525	-	3206	-	-	-
Low	-	-	10	0	9	0	2	2	-	1	-	-	-
Mean	-	-	7490	542	15283	1106	40926	85	-	317	-	-	-
Median	-	-	2935	19	3415	25	10186	54	-	114	-	-	-
Salt													
	pH	Temp.	Na^+	K^+	Ca^{2+}	Mg^{2+}	Cl^-	HCO_3^-	NO_3^-	SO_4^{2-}	HS^-	F^-	PO_4^{3-}
High	8.1	-	123000	30000	18700	45000	204000	30	-	17500	-	7	-
Low	6	-	10651	15	100	10	18980	3.8	-	250	-	4	-
Mean	-	-	-	-	-	-	-	-	-	-	-	-	-
Median	-	-	-	-	-	-	-	-	-	-	-	-	-
Argillaceous (Clay/Shale)													
	pH	Temp.	Na^+	K^+	Ca^{2+}	Mg^{2+}	Cl^-	HCO_3^-	NO_3^-	SO_4^{2-}	HS^-	F^-	PO_4^{3-}
High	12	242	126957	1575	37300	22632	204000	8150	-	15000		0.51	-
Low	1.0404	97	3	3	1	1	6	4.56	-	1		0.51	-
Mean	7.39	142.32	19099	135	3088	708	35830	964	-	1270		0.51	-
	7.3	135	12143	48	1140	290	21588	448	-	682		0.51	-

Number of Data Points for the Argillaceous (clay/shale) Section

Parameter	**# of Data Points**	**Parameter**	**# of Data Points**
pH	710	HCO_3^-	762
Temp.	26	NO_3^-	0
Na^+	772	SO_4^{2-}	721
K^+	167	HS^-	0
Ca^{2+}	787	F^-	1
Mg^{2+}	776	PO_4^{3-}	0
Cl^-	794		

Notes:

1. The granite data are from Frape et al. (2003). These data are for sites in Canada, Sweden, Finland, Russia, Western Europe, and the UK.
2. Salt data are from Tables 21 and 23 of Bryan et al. (2011), Stein and Krumhansl (1987), and Deal et al. (1995).
3. Shale data are from the U.S. Geological Survey National Produced Waters Geochemical Database V2.0 (Blondes et al. 2014). The data are for U.S.

geological formations only, and are based on produced waters. They span a range of dates, with the most recent being in 1980.

Salt Repository Disposal Environments

The main characteristics of salt repositories are summarized from the literature review compiled by Bryan et al. (2011). Creep closure is expected to seal the drifts or boreholes used to emplace radioactive waste, and the creep closure rate depends on the lithostatic pressure and the temperature. A typical salt repository is considered impermeable due to viscoplastic behavior. Water content depends on the salt formation. Bedded salt has brine content that ranges from a few tenths to a few weight percent. Domal salt has less brine, from a few thousandths to tenths of a weight percent (Bryan et al. 2011). The brine exists in the intragranular spaces, within fluid inclusions, and as structural water in hydrous minerals (clay minerals, gypsum, and polyhalite). Typical brines contain sodium and chloride ions, and sometimes significant amounts of dissolved magnesium (Table 2). Overall, salt formations provide little or no pH or redox buffer capacity, and the redox conditions adjacent to the waste package are expected to be controlled by reactions involving package materials (including radiolysis products), controlled by the availability of brine, and limited oxygen fugacity. Waste heating of the salt may cause brine migration which could change brine availability.

Argillaceous Disposal Environments

Argillaceous (clay/shale) host media can be either saturated or unsaturated. Argillaceous rocks are characterized by low permeability, reducing conditions, presence of sulfide and organic matter, with variable compositions of pore water. These waters range from dilute sodium bicarbonate to medium-strength calcium-sodium-chloride-sulfate type (Table 2).

3. Survey of corrosion rates and mechanisms, and corrosion products for stainless steels AISI 304L and 316L

Alloys 304L and 316L are the most common materials in DPC shells and basket structural components. A schematic representation of the corrosion drivers and corrosion products for Fe-based alloys (carbon and stainless steels) is shown in Figure 3.

Redox Effects

The redox potential of the system, particularly the availability of dissolved oxygen, controls the corrosion pathway and formation of corrosion products. Both oxic and anoxic conditions are expected during the postclosure performance period of a repository. General and localized corrosion proceed faster in oxic conditions, but oxic conditions are expected to exist early during the evolution of repositories in low-permeability host media or backfill/buffer materials.

Under oxic conditions cathodic reactions on metallic surfaces are dominated by the reduction of dissolved oxygen (Figure 3) (Rebak 2011). Major solid corrosion products of the Fe-based alloys (carbon and stainless steels) under oxic conditions are iron oxy(hydr)oxides goethite (FeOOH), lepidocrosite (Fe_2O_3), hematite (Fe_2O_3), maghemite (Fe_2O_3), and magnetite (Fe_3O_4) (Jack and Wilmott 2011).

Most DPC direct disposal concepts (and other disposal concepts as well) are expected to return to anoxic or anaerobic conditions shortly after backfilling and closure (Bryan et al. 2011). This is especially true for low-permeability, water saturated host media that contain reducing minerals (e.g., pyrite) or natural organic matter. Under reducing conditions, water acts as an electron acceptor for metallic iron, and the cathodic reaction is controlled by hydrogen evolution (Figure 3) (Rebak 2011). The ubiquity of water as a possible electron acceptor is one reason that steels continue to corrode, albeit slowly, at anoxic aqueous conditions. Under anoxic conditions, siderite ($FeCO_3$), iron hydroxide $Fe(OH)_2$ and magnetite (Fe_3O_4) are the predominant corrosion products of the Fe-based alloys (Jack and Wilmott 2011). During anaerobic microbial assisted corrosion a different assemblage may form including siderite, amorphous iron-sulfide (FeS), mackinawite (FeS), greigite (FeS), and pyrite (FeS_2) (Jack and Wilmott 2011).

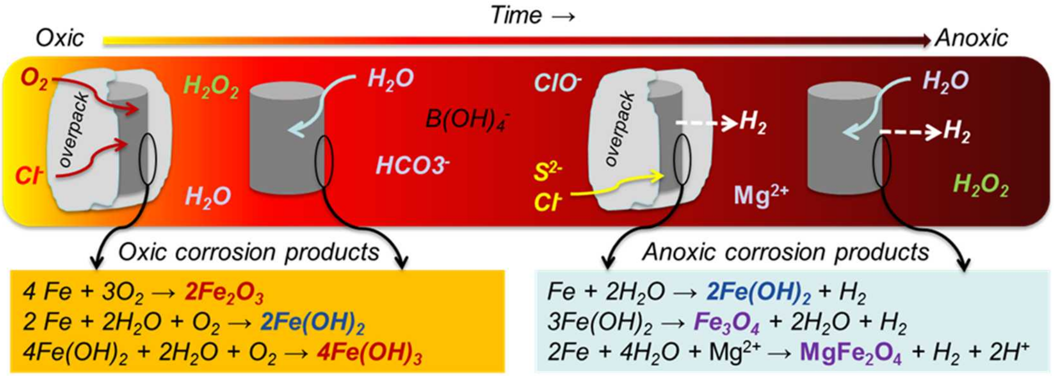


Figure 3. Time-dependent chemical corrosion drivers for Fe-based materials (carbon steel and stainless steel)

While steel corrosion is generally anticipated to be slower under anoxic conditions, environmental factors such as sulfide concentration or microbial activity may result in rapid corrosion even under anoxic conditions. For instance, a qualitative soil corrosivity classification has been developed based on the soil oxidation-reduction potential (ORP) by (Jack and Wilmott 2011). They observed a increase in corrosion with increasingly reducing (lower ORP) conditions; no corrosion at ORP>400 mV, slight corrosion at 200 to 400 mV, moderate corrosion at 100 to 200 mV, and severe corrosion at ORP<100 mV. This was attributed to the effects of anaerobic microbial activity in the anoxic soils. The relative importance of microbial activity in repository settings may vary with the host rock. It is generally believed to be suppressed in highly compacted swelling clay backfill materials such as those used in granite repository concepts (Kwong 2011). However, tests in the Opalinus Clay at Mont Terri (e.g., Wersin et al. 2011; Vinsot et al., 2014) have shown high rates of microbial activity when electron donors (H_2 or incidental organic matter) were introduced via boreholes; sulfate was the electron acceptor. Hence, the potential for microbially mediated corrosion cannot be ignored.

Effects from pH and Chemical Composition on Corrosion Mechanisms and Rates

As discussed above, pH and composition of the aqueous phase are major controls on corrosion rates and mechanisms. Typically, soil pH falls within the range 3.5 to 10. Corrosion of Fe-based alloys increases significantly as pH falls below 4 (Jack and Wilmott 2011). Figure 4 illustrates the dependence of steel corrosion rates on pH and resistivity of ground water, for steel pipes buried in near-surface soils where redox conditions are typically oxidizing. Groundwater resistivity is a surrogate for greater salt content. The effects of pH and salt content are similar under anoxic conditions, but uniform corrosion rates are expected to be lower (compared to Figure 4).

Under anoxic conditions, localized corrosion modes include hydrogen blistering, hydrogen-induced cracking, stress-oriented hydrogen-induced cracking, and sulfide stress cracking (Elboujdaini 2011). Hydrogen gas is one of the corrosion products of Fe-based alloys in anoxic conditions (Figure 3). Hydrogen embrittlement, induced blistering, and cracking occur due to the evolution of atomic hydrogen at the surface, followed by its diffusion into the steel. Once entrained within the steel, the hydrogen accumulates in hydrogen traps (e.g., around inclusions), leading to localized pressure increase within the material (Elboujdaini 2011). Sulfide stress cracking is a variety of hydrogen-induced cracking, and is usually localized in weld zones (Elboujdaini 2011). It can occur in mildly corrosive media at temperatures below 90°C. Sulfide reaction with ferrous iron:

$$H_2S + Fe^{2+} \rightarrow FeS + 2H^0 \tag{8-1}$$

produces atomic hydrogen, and dissolved sulfide hinders hydrogen recombination reaction at the corroding surface (combining two atomic H^0 into gas H_2). Abundant atomic hydrogen easily diffuses into steel (Elboujdaini 2011).

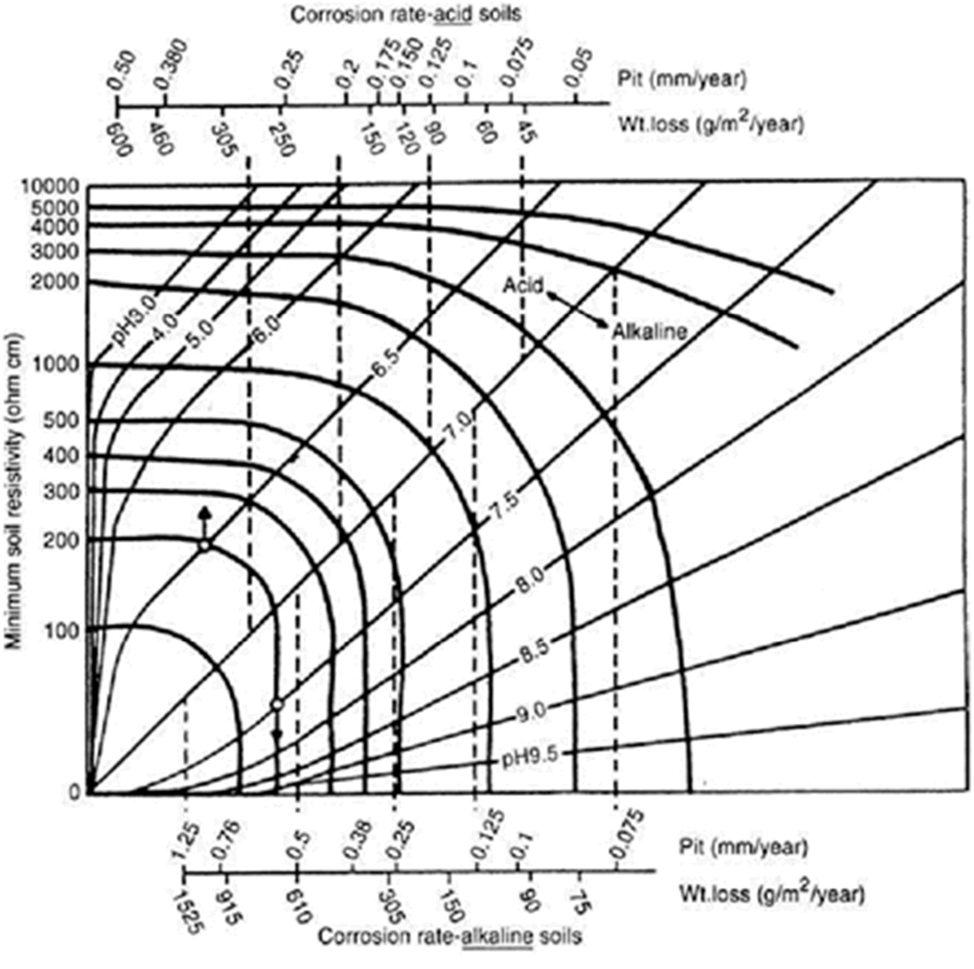


Figure 4. Nomogram relating soil resistivity, pH, and corrosion rate for steel pipe in soil (from Jack and Wilmott 2011; original source King 1977)

Radiation Flux Effects

Radiation exposure has a strong effect on the corrosion environment. Container corrosion rates, particularly in anoxic repository settings, could be significantly increased by radicals produced by radiolysis. The following short discussion on radiolysis is a summary from the recent report by Buck et al. (2012).

The redox conditions in a waste repository environment are expected to evolve due to the time-dependent generation of radiolytic redox active species (both oxidants and reductants), and the corrosion of Fe-bearing canister materials. In the presence of water vapor or a thin-film of water, the γ-radiation from SNF triggers complex radiolysis reactions (about 100 have been identified). Some of the products of these reactions include hydroxide (OH•) and hydrogen (H•) radicals, oxygen ions (O_2^-), aqueous electrons (e^-_{aq}), hydrogen peroxide (H_2O_2), hydrogen gas (H_2), and the secondary radiolysis product, oxygen (O_2). These species are highly reactive, and are expected to increase both the degradation rate of the SNF, and the corrosion rate of the container. Hydrogen peroxide (H_2O_2) can significantly increase pitting rates of stainless steel (see below) and is expected to be the predominant oxidant for SNF under anoxic conditions (Buck et al. 2012).

Survey of Corrosion Rate Data for Stainless Steel

Stainless steel types 304/304L and 316/316L are the most common alloys used in the construction of DPCs and their internal components. Type 304 is a chromium-nickel alloy, and type 316 is a chromium-nickel alloy containing 2 to 3% molybdenum.

Uniform Corrosion of Stainless Steel – Under oxic, alkaline conditions, and the water chemistry typical of a clay repository (no added chloride) uniform corrosion rates for 316L vary from <0.1 $\mu m\ yr^{-1}$ at 30ºC to 0.2-0.8 $\mu m\ yr^{-1}$ at 80ºC (Kursten et al. 2004). Estimated corrosion rates under anoxic conditions are from 0.001 $\mu m\ yr^{-1}$ to 0.1 $\mu m\ yr^{-1}$ over the same temperature range (Kursten et al. 2004). A proposed long-term general corrosion rate for stainless steels under anaerobic conditions is <0.01 $\mu m\ yr^{-1}$ (Kursten et al. 2004).

Localized Corrosion of Stainless Steel – Experimental testing of indicated no pitting of 316L in alkaline solutions containing up to 100 $g\ L^{-1}$ chloride at room temperature (Kursten et al. 2004). When the concentration of chloride was decreased to 50 $g\ L^{-1}$ the critical pitting temperature is increased to 45ºC (Kursten et al. 2004). Similar chloride concentration threshold behavior was observed for pitting of 304L: pitting was observed at 60ºC with >50 $g\ L^{-1}$ chloride (Kursten et al. 2004). Crevice corrosion of 304L is observed at 80ºC and background chloride concentrations of 20 $g\ L^{-1}$ or greater; and no crevice corrosion is observed at 40ºC and chloride concentrations up to 20 $g\ L^{-1}$ (Kursten et al. 2004). Pit initiation testing for 304/304L and 316/316L indicates that pitting is variable (Table 3). The oxidative history of the sample was associated with a large difference in the number of pits for the 316/316L alloys, but not for 304/304L.

Stress corrosion cracking was observed in an unstressed sample of 304L stainless steel that had been aged in cementitious material containing 100 $g\ L^{-1}$ chloride for 2 years. Additional testing in alkaline solutions indicated that increased chloride (17.7 g/L) and thiosulfate ($S_2O_3^{2-}$ at 3.4 g/L) increased both pitting and stress corrosion cracking of the 316L and 304L alloys (Kursten et al. 2004).

In aggressive environments (e.g., 45% MgCl or 26% NaCl), stress corrosion cracking is observed to take place within hours to days. Cracking is observed in less than 3 hours in magnesium chloride solution at 155°C, and after 48 to 72 hours in sodium chloride tests at 102 and 200°C (Streicher and Grubb 2011).

Table 3. Pit initiation in stainless steels 304/304L and 316/316L exposed to 0.1N NaCl at 25°C (from Streicher and Grubb 2011)

	Element %						Pits per cm^2 (Note 1)	
Alloy	**Cr**	**Ni**	**Mo**	**C**	**N**	**Si**	**Clean** ^{2}	**Passivated** ^{3}
304	18.45	8.90		0.063		0.58	3.4	2.2
304L	18.30	11.02		0.020	0.033	0.37	1.6	2.8
316	17.93	13.50	2.47	0.031		0.31	0.46	15.8
316L	17.71	11.17	2.44	0.02	0.032		0.17	29.0

Notes:

1. Pits produced by anodic polarization.
2. Surface cleaned in nitric-hydrofluoric-hydrochloric HNO_3-HF-HCl mixture prior to the experiment.
3. Surface passivated in nitric acid HNO_3 with potassium dichromate $K_2Cr_2O_7$.

4. Research Needs for Predicting Stability of DPC Materials in Disposal Environments

The neutron absorber materials in current DPCs are largely aluminum-based and are not anticipated to survive long in a breached, flooded waste package. The aluminum-based neutron absorber will actually act as a sacrificial anode with respect to the steel. Borated stainless steel, used in a few DPC packages, is anticipated to have a longer lifetime. Corrosion of borated stainless steel under reducing conditions has not been explored, and further testing is needed to assess this material if it will be used in future canisters.

Criticality analysis (Section 4) has shown that the basket degradation case is the most reactive configuration, but might be avoided if stainless steel basket structures maintain the SNF configuration during the postclosure repository performance period (e.g., up to 10,000 years). A rough estimate of the expected loss of thickness of a stainless steel plate or sheet, after 10,000 years exposure to ground water, is 0.01 to 1 mm (1-sided surface retreat). This estimate is based on the following assumptions:

- Anoxic conditions, non-corrosive water composition, and pH >4
- Estimated stainless steel corrosion rates for anoxic conditions are 0.001 to 0.1 $\mu m\ yr^{-1}$ over the temperature range 30 to 80°C (Kursten et al. 2004)
- Localized corrosion (e.g., pitting) may occur but does not cause loss of structural integrity
- Crevice corrosion and stress corrosion cracking do not occur, or if they do occur damage to the basket does not impact structural integrity
- Microbially influenced corrosion does not occur
- Hydrogen that evolves at corroding surfaces diffuses away and does not accumulate in the uncorroded steel, so the extent of any hydrogen-embrittlement or hydrogen-induced cracking is not significant

- Radiolysis products (e.g., H_2O_2) predominantly react with UO_2 in the SNF, or recombine, and do not react with basket materials to any significant extent because the cladding is mostly intact

Using this estimate, a stainless steel basket structure could maintain the SNF configuration for 10,000 years (e.g., in the event of early breach of the disposal overpack).

Some of these assumptions should be tested experimentally. Investigation should focus on container-specific corrosion rates, and testing or analysis to evaluate the assumptions used for this estimate of basket lifetime. Corrosion processes are complex, and there are no existing mechanistic process models that could more reliably predict corrosion rates. Whereas some basket designs have relatively thin-wall stainless steel tubing or sheets, under reducing conditions general corrosion rates are generally slow. However, within a waste package radiolysis may create oxidizing conditions that could locally increase corrosion rates. While stainless steels will pit in reducing, saline ground waters it is not clear that this will structurally weaken the basket. Crevice corrosion and SCC of basket materials may be more important in limiting basket lifetime. Crevice corrosion may occur at any contact, while SCC may occur in weld zones or areas of high loading. Another potential factor may be hydrogen embrittlement associated with anoxic metal corrosion reactions or by radiolytic breakdown of water.

Key areas of uncertainty that should be addressed by future investigations include the evolution of disposal environments through time:

- Changes in redox conditions, temperature, hydrogen buildup/diffusion, interactions between products of corrosion (e.g., Fe^{2+}) and the clay buffer
- Diffusion of corrosion-active species (e.g., chloride or sulfide) through buffer material, and potential effects on long-term corrosion rate
- Feedbacks between H_2 production and transport
- Likelihood of localized corrosion in basket materials
- Radiation effects on the corrosion environment, particularly in anoxic repository settings, from radicals produced by radiolysis (e.g., H_2O_2)
- Competition for oxidizing radicals between UO_2 and container materials

and aspects of container design and fabrication that can affect corrosion:

- Effects of thermal treatments and welding of the basket
- Metallurgical modifications due to long-term radiation flux or thermal aging

Prospective Overpack Materials

For DPCs, barrier functions will be assigned to the overpack because the 1.27 to 1.59 cm (1/2 to 5/8 inch) shell is not expected to provide structural strength for handling and emplacement, or corrosion resistance to isolate the waste for the required repository performance period. To be an effective barrier, the overpack/waste package system must resist damage during handling and emplacement. Additionally, for clay and salt repositories after backfilling, pressures will rise to those of the host rock with magnitude between hydrostatic and lithostatic. Any overpack must resist crushing under such loading conditions.

There are two general strategies for overpack material selection: corrosion allowance material and corrosion-resistant material. Corrosion allowance has been used in European clay/shale and salt repository designs, with selection of thick-wall carbon steel waste packages. A thick wall can resist crushing until the overpack is degraded by corrosion. Although carbon steel corrodes relatively rapidly via general corrosion, it is not susceptible to pitting or crevice corrosion. Hence, canister degradation rates can be reliably calculated. Containment penetration times for thick carbon steel overpacks or waste packages are typically estimated to be on the order of 10,000 years, which can be consistent with repository performance objectives if other engineered and natural barriers continue to perform isolation functions. Accordingly, the use of corrosion allowance materials may be limited to salt and other repository concepts where the natural system is an effective long-term isolation barrier.

Corrosion-resistant materials such titanium or copper, and including stainless steels, can produce longer estimates containment lifetime. Materials such as titanium and stainless steel passivate under the environmental conditions being considered, forming a resistant surface oxide layer leading to very low general corrosion rates. Passive materials are by their nature susceptible to localized corrosion, or disruption of the passive layer with rapid local penetration (e.g., pitting, crevice corrosion, or stress corrosion cracking). Copper is a special case because it does not passivate. Rather, the metal is thermodynamically stable in the presence of water, and will not corrode under anoxic conditions in pure water. However, it will corrode in natural anoxic groundwater systems due to the presence of sulfide, with which it reacts to form Cu-sulfides. Among corrosion-resistant materials, only copper is currently being considered for a waste package (although other corrosion-resistant materials have been considered in the past). Because corrosion-resistant materials are more expensive than carbon steel, they would likely be used in a relatively thin layer as part of a DPC disposal overpack. Structural requirements of the overpack could then be met by another, potentially thicker layer (e.g., stainless steel).

In European repository concepts using corrosion allowance materials, criticality concerns are addressed using two strategies. In French, Swiss, and Belgian clay repository designs, the SNF capacity is restricted (e.g., 4 PWR size, or typically 9 BWR assemblies) which can be small enough to prevent criticality. For the German POLLUX fuel rod consolidation is proposed, which significantly decreases the amount of moderating water that can combine with the fuel. DPCs have much greater capacity, and contain intact fuel assemblies, so these strategies are not applicable. Hence, an overpack lifetime of 10,000 years could be needed for DPC disposal overpacks, to exclude ground water. Note that the 10,000 year time requirement is a regulatory one (Hardin and Howard 2013). Carbon steel corrosion rates under anoxic conditions are well known and can be used to calculate the needed thickness.

For overpacks constructed of a corrosion-resistant material, work by the European research groups has focused mainly on copper. Stainless steel and Ni-based alloys are susceptible to localized corrosion and potentially, rapid penetration. Moreover, under anoxic conditions, corrosion will occur via reduction of water, producing hydrogen. Both stainless steel and low-iron nickel alloys are susceptible to hydrogen uptake and embrittlement when this reaction occurs. Corrosion reactions with hydrogen sulfide present in reducing groundwaters can also lead to hydrogen embrittlement of steel and Ni-based alloys. Titanium is not affected by hydrogen sulfide, unless it is galvanically coupled to a less noble metal (e.g., a steel alloy). Copper is also not affected by hydrogen embrittlement. If nickel alloys or stainless steels are proposed for an overpack material, then corrosion rates, and pitting penetration rates for these materials would be determined for each relevant environment.

Other materials currently under consideration for engineered barriers, which may include disposal overpacks, include: gray cast iron, 1018 carbon steel, 4130 alloy steel, 2.25Cr-1Mo type 304 and 316 stainless steels, Monel 400, Incoloy 825, Inconel 625, Hastelloys C-4 and C-22, and

Ti (Grades 2, 7, 12, 16 and 29) (Rebak 2011). Some corrosion studies for two types of prospective overpack materials (carbon steel and Hastelloys C-4 and C-22) are summarized below.

Corrosion of Carbon Steels

Corrosion of a waste container is expected to depend mainly on the immediate geochemical environment of the container. For example, if a backfill material is used (e.g., bentonite clay backfill in a granite repository) then geochemical conditions within the backfill will drive the corrosion of waste package materials. Relevant corrosion data for two types of carbon steel (BS4360 grade 43A and TStE 355) are discussed here. For the former, data are presented for exposure to bentonite systems, while for the latter, data are reported for exposure to brine. Experiments were completed to measure the rates of carbon steel (BS4360 grade 43A) corrosion in a bentonite clay slurry, compact bentonite, and pore water simulants. The measured uniform corrosion rates for carbon steel ranged depending on temperature and whether it was exposed to the compact media, slurry, or homogeneous (pore water) phases. The observed corrosion rates varied with time. Fresh carbon steel surfaces corroded rapidly, with the rate increasing with increasing temperature. The initial rate was around 25 to 30 $\mu m\ yr^{-1}$ for bentonite slurries at 30 and 50ºC. This high rate was sustained for a short time, and dropped to 1.5 to 4 $\mu m\ yr^{-1}$ after ~3,000 hours of reaction for all studied systems (Smart et al. 2006). In the homogeneous systems the long-term (>2,500 days) carbon steel general corrosion rates in anoxic alkaline conditions at 30, 50 and 80°C were <0.1 $\mu m\ yr^{-1}$ (Smart et al. 2004). The decrease in corrosion rate is due to the development of oxide layers, namely ferrous hydroxide at 30ºC and magnetite at 50 and 80ºC (Smart et al. 2004).

Carbon steel TStE 355 corrosion rates vary widely depending on the chemical conditions of the experiment. From *in situ* studies where the amount of brine was limited, an estimated general corrosion rate was <0.1 $\mu m\ yr^{-1}$ (90 and 170ºC) (Kursten et al. 2004). In the laboratory experiment with excess brine, the resulting corrosion rates were 70 $\mu m\ yr^{-1}$ (90ºC) and 199 $\mu m\ yr^{-1}$ (170ºC) in magnesium-rich brines, and 5 $\mu m\ yr^{-1}$ (90ºC) and 46 $\mu m\ yr^{-1}$ (170ºC) in sodium chloride brines (Kursten et al. 2004). The predominant corrosion mode for carbon steel is uniform corrosion, but sometimes stress corrosion cracking can occur when the environmental conditions allow for development of a passivating layer, for example in the presence of anodic inhibitors: nitrates, hydroxides, carbonates, or phosphates (Parkins 2011).

Survey of Corrosion Rate Data for Hastelloys C-4 and C-22

The corrosion behaviors of stainless steels, nickel-based alloys (Hastelloys C-4 and C-22), Ti99·8-Pd and copper-based materials in rock salt, granite and clay environments were assessed in the laboratory by Smailos et al. (2004). The corrosion results for Hastelloys C-4 and C-22 are summarized below. Overall, the study concluded that the most promising materials for disposal in granitic formations are Hastelloy C-22, Cu and Cu-Ni-alloys. For clay/shale formations, the most important candidate materials for thin-walled containers are stainless steels, nickel-based alloys (Hastelloys C-4 and C-22), and Ti99·8-Pd (Smailos et al. 2004).

Corrosion studies on the nickel-based alloys Hastelloy C-4 and C-22 in rock salt, granite, and clay environments indicate that these materials exhibit excellent general and local corrosion resistance. For anoxic conditions in granitic environments (Cl^- up to 50,000 mg/L, temperature up to 90°C) Hastelloy C- 22 exhibits the highest corrosion resistance among all tested materials. It is resistant to pitting corrosion, crevice corrosion, stress corrosion cracking and microbially influenced corrosion. For clay/shale repository conditions (100 to 50,000 mg/L Cl^-, at 16°C and up to 140°C) Hastelloys C-4 and C-22 show slight crevice corrosion under severe test conditions (oxidizing, 140°C, $Cl^- > 20,000$ mg/L) (Smailos et al. 2004).

Under aerobic (oxic) conditions at 140°C, Hastelloys C-4 and C-22 are resistant to general and pitting corrosion. However, signs of pitting were observed in some tests with high Cl^- (> 20,000 mg/L) (Smailos et al. 2004). The addition of hydrogen peroxide (H_2O_2) to the solutions (to mimic the effect of potential radiolysis products) did not affect the corrosion behavior of Hastelloys C-4 and C-22 in both oxic and anoxic conditions at 90°C (Smailos et al. 2004).

Summary and Application to DPC Inventory

The foregoing discussion shows that stainless steel corrosion rates may be low enough to sustain DPC basket structural integrity for performance periods of as long as 10,000 years, especially in reducing conditions. Uncertainties include basket component design, disposal environment conditions, and the in-package chemical environment including any localized effects from radiolysis.

Published data briefly reviewed above support an observation that prospective disposal overpack materials exist for most disposal environments, including both corrosion allowance and corrosion resistant materials. Whereas the behavior of corrosion allowance materials is understood for a wide range of corrosion environments, demonstrating corrosion resistance could be more technically challenging and require environment-specific testing.

Not all DPCs have stainless steel basket structures, as noted for the Site C canisters analyzed in Section 4. The following sections describe a preliminary screening of the existing inventory of DPCs and other types of canisters, according to the type of closure, whether they can be readily transported, and what types of materials are used in basket construction.

Cask Closure

Storage and transportation casks can be broadly subdivided into two categories, those in which UNF fuel is stored in thin-wall, welded metal canisters and those in which fuel is stored in metallic canisters with bolted closures. DPCs are welded metal canisters that can be transferred between overpacks for storage, transportation and possibly disposal.

Bolted-closure systems are typically referred to as “casks” because they are massive and self-shielding, and cannot be inserted into overpacks for other purposes such as disposal (and in some cases, transportation). Bolted-closure systems may also be referred to as “bare fuel casks” because they are designed for fuel retrieval (as bare assemblies) and cask reuse. Regulatory requirements for bolted closures include periodically monitoring the inerting gas pressure over the life of the cask, whereas welded canisters are known to be leak-tight. Metal gaskets are typically used to aid in the sealing of bolted lids, and must be replaced if a leak occurs. Gamma and neutron shielding are typically integral to the bolted container.

A bolted system could be suitable for disposal if a disposal overpack with appropriate dimensions and postclosure performance is available. Bolted closures must be monitored and maintained and thus would not be suitable for direct disposal without an additional, permanent containment envelope. Since there are relatively few existing dry storage systems in the U.S. with bolted closures, and they can be readily opened to retrieve the fuel for disposal, they are not considered in the canister screening exercise described below (Section 8.6.5).

Transportability

For purposes of this screening the existing DPC inventory is divided into canisters designed for storage and transportation and those that are considered to be storage only. Canisters are considered to be transportable if they have a 10CFR71 certificate of compliance, or if they are new designs that have not completed the 10CFR71 licensing process. It may be possible in the future to license some older storage-only canisters for transportation, but that is beyond the scope of this analysis.

Repository Degradation Susceptibility

As shown in Section 4, structural integrity of the fuel basket (e.g., for up to 10,000 years) greatly increases the fraction of analyzed DPCs that could remain subcritical after flooding. Basket integrity will be determined by the susceptibility of the basket materials to corrosion. As discussed above, stainless steel is the most degradation resistant material used in canister construction, and may retain sufficient structural integrity in the repository environment after exposure to ground water. By obviating the basket degradation reactivity scenario, stainless baskets could prevent criticality. For this analysis, stainless steel is assumed to be the only material in existing DPC that could perform in this manner. A working definition of degradation susceptibility for basket materials is provided below.

Basket Designs and Structural Components

There are two major types of baskets used in DPCs: the “tube-and-spacer-disk” design (Figure 5) and the “egg-crate” design (Figure 6). The tube-and-disk design consists of a series of fuel tubes (also called guide sleeves) which maintain the position of each fuel assembly. The fuel tubes are held in place by mechanical coupling to a set of disks, which are secured to the canister body by tie rods. The disks maintain the tube spacing and provide heat transfer to the canister shell. Some of the disks may be fabricated from aluminum or other materials to aid heat transfer. The egg-crate designs typically comprise fuel support tubes or cells made from tubing or plates welded together. The entire basket is connected to the canister by supports attached at the basket periphery. Egg crate designs may also incorporate interstitial material between the fuel tubes in order to improve heat conduction out of the basket.

For screening of tube-and-disk canisters the materials of construction of the spacer disks, support rods, and fuel tubes were examined. For screening the egg-crate canisters only the fuel tube materials were considered, eliminating some of the support design details because it seems likely that the supports could fail without causing a structural failure of the basket.

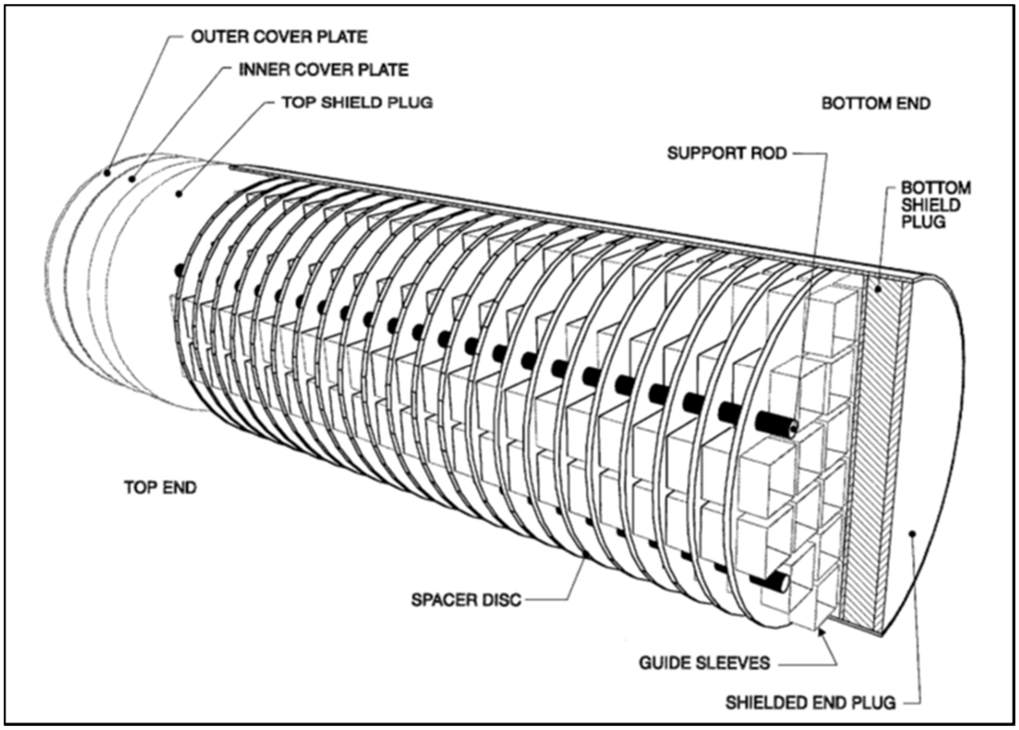


Figure 5. Schematic cutaway of the FO/FC-DSC manufactured by Transnuclear, an example of tube-and-disk basket design

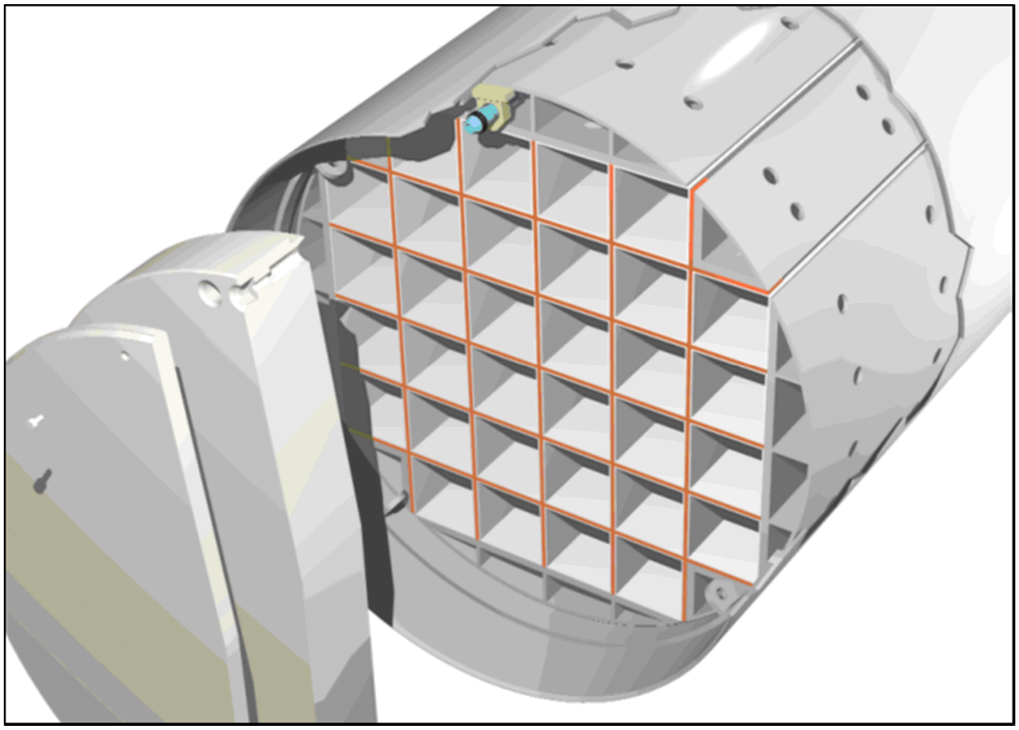


Figure 6. Schematic cutaway of the DSC-32PTH manufactured by Transnuclear, an example of egg-crate basket design

Canister Inventory and Screening

Screening to determine the fraction of canisters or fuel assemblies that may be stored in disposable containers (as defined) begins with canister inventory, PWR and BWR assembly counts, and other data from a table compiled by an industry newsletter publication (Ux Consulting 2014). The materials of construction of each canister were extracted from licensing documents (Transnuclear 2002, 2012; McLean 1990; NAC 2003, 2004, 2010; NRC 2014; Holtec 2010; Energy Solutions 2002, 2003). The compilation for existing DPCs is presented in Table 4. Based on the criteria discussed above the canisters were grouped into the following categories:

- Transportable canisters without degradation-susceptible components
- Transportable canisters with degradation susceptible-components
- Non-Transportable canisters without degradation-susceptible components
- Non-Transportable canisters with degradation-susceptible components
- Bolted casks

Degradation susceptibility is defined here to mean non-stainless steel structural components. Thus, DPCs that have carbon steel spacer disks (including plated steel), or aluminum-based (e.g., Metamic®) basket structures, would be degradation-susceptible. DPCs that contain non-stainless materials that are not used in structural applications (e.g., thermal shunts) would not be susceptible as long as the structure is stainless steel. This categorization does not imply that stainless steel degradation could not occur. It is intended to show which canisters could have sufficient structural lifetime in relatively fresh ground waters, that the criticality analysis could consider the loss-of-absorber scenario but not necessarily the basket degradation scenario.

The results of the screening process are shown in Figures 8-7 through 8-9, and Table 5. Approximately 2/3 of the overall inventory of storage casks and canisters are considered transportable with basket structural components made from stainless steel, while 6% are transportable but with non-stainless components. The remaining 25% consists of storage-only canisters and bolted casks. The fraction of BWR assemblies in transportable canisters with stainless steel basket components (83% of all BWR fuel), is greater than the fraction of PWR assemblies (65% of all PWR fuel).

It should be reiterated that these screening results are based on assumptions that stainless steel is the best existing basket material for disposability, that component thickness (e.g., guide sleeves, Figure 5) is sufficient to sustain structural integrity, and that basket supports and interstitial materials (Figures 8-5 and 8-6) are not critical to basket structural integrity.

Table 4. Domestic inventory of UNF in dry storage, by canister type, transportability, and susceptibility to degradation in disposal environments

Canister/Cask Name	**Closure**	**Reactor Type**	**Number of Canisters Loaded**	**Number of Assemblies in Storage**	**Transportable**	**Susceptible to Repository Degradation**	**Reference (see notes)**
TN-32	Bolted	PWR	63	2016	Not Included	Not Included	Not Included
TN-40		PWR	35	1400			
TN-68		BWR	68	4624			
V/21&X33		PWR	26	558			
MC-10		PWR	1	24			
NAC-I28		PWR	2	56			
DSC-24PTH	Welded	PWR	27	648	Yes	No	1
DSC-32P		PWR	24	768	No	No	2
DSC-32PT		PWR	76	2432	Yes	No	1
DSC-32PTH		PWR	98	3136	Yes	No	1
DSC-61BT		BWR	129	7869	Yes	No	1
DSC-61BTH		BWR	51	3111	Yes	No	1
MPC-24		PWR	20	480	Yes	No	3
MPC-24EEF		PWR	34	790	Yes	No	3
MPC-32		PWR	272	10324	Yes	No	3
MPC-68		BWR	300	20400	Yes	No	3
MPC-HB		BWR	6	390	Yes	No	3
TSC-37		PWR	23	851	Yes	Yes	4
DSC-24P		PWR	135	3240	No	Yes	2
DSC-24PHB		PWR	48	1152	No	Yes	2
DSC-24PT1		PWR	18	395	Yes	Yes	5
DSC-24PT4		PWR	33	792	Yes	Yes	1
DSC-52B		BWR	27	1404	No	Yes	2
DSC-7P		PWR	8	56	Yes	No	6
DSC-FO/FC		PWR	22	493	Yes	Yes	5
CY-MPC		PWR	43	1019	Yes	No	7
MPC-		BWR	5	333	Yes	No	7
MSB		PWR	58	1392	No	Yes	8
TSC-24		PWR	236	5562	Yes	No	9
W74		BWR	8	441	Yes	Yes	10
Yankee-DPC		PWR	16	533	Yes	No	7

Notes:

1. Transnuclear (2012)
2. Transnuclear (2002)
3. Holtec (2010)
4. NAC (2010)
5. NRC (2014)
6. McClean (1990)
7. NAC (2003)
8. Energy Solutions (2002)
9. NAC (2004)
10. Energy Solutions (2003)

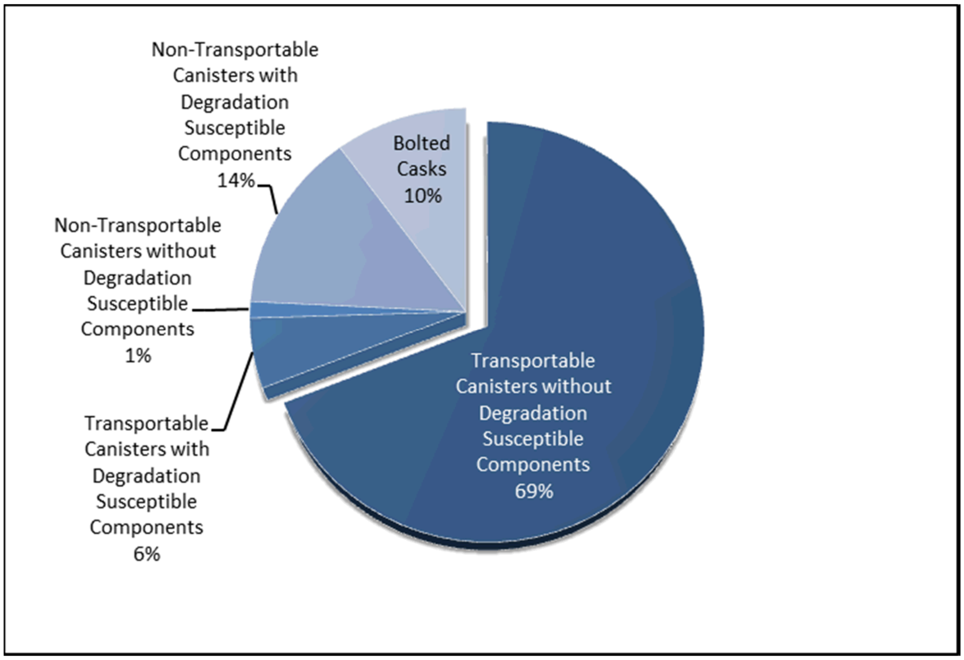


Figure 7. Fractional representation of the total number of DPCs and bare fuel casks by canister type and material susceptibility

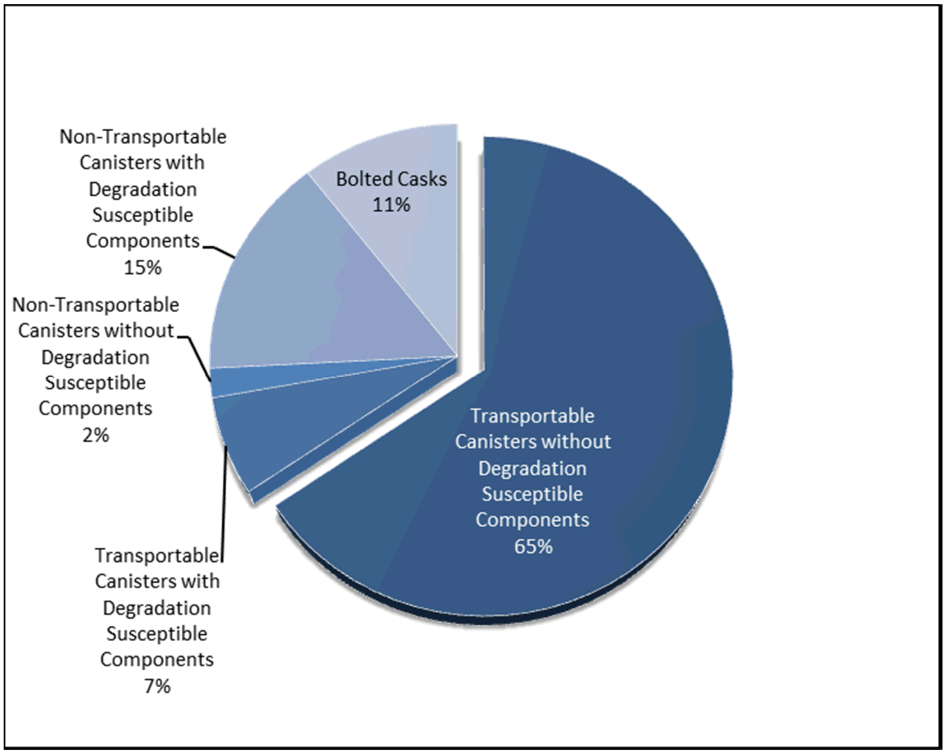


Figure 8. Fractional representation of PWR fuel assemblies by canister type and material susceptibility

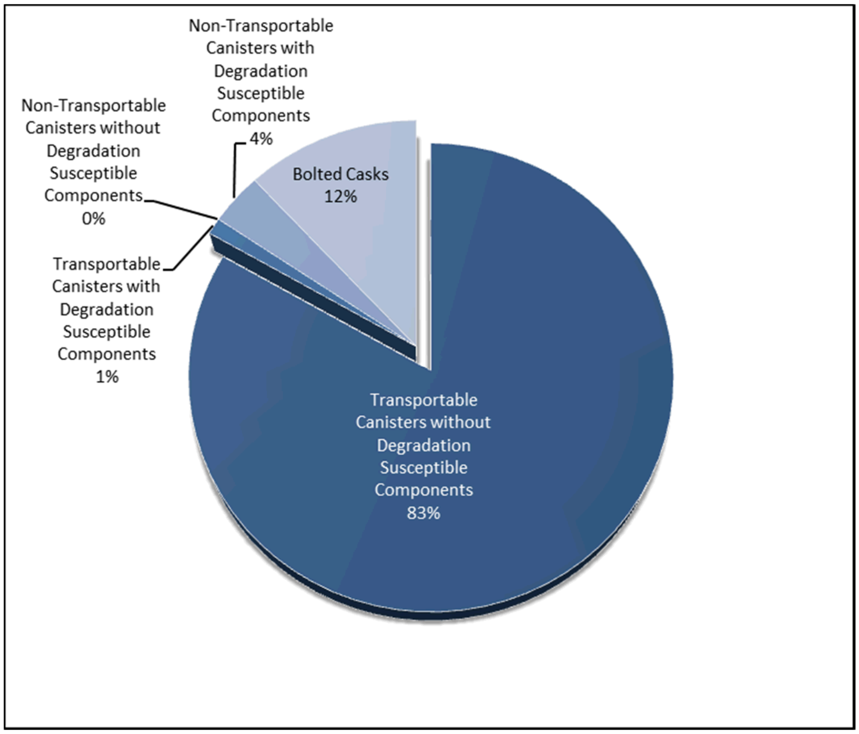


Figure 9. Fractional representation of BWR fuel assemblies by canister type and material susceptibility

Table 5. Summary of the distribution of canisters and fuel types, by canister type and material susceptibility

Canister Classification	**# of Canisters in Storage (July, 2014)**	**# of PWR Fuel Assemblies in Storage (July, 2014)**	**# of BWR Fuel Assemblies in Storage (July, 2014)**
Transportable Canisters without Degradation-Susceptible Components	1,321 (69.1%)	24,980 (65.5%)	32,103 (83.2%)
Transportable Canisters with Degradation-Susceptible Components	104 (5.4%)	2,531 (6.6%)	441 (1.1%)
Non-Transportable Canisters without Degradation-Susceptible Components	24 (1.3%)	768 (2.0%)	0 (0%)
Non-Transportable Canisters with Degradation-Susceptible Components	268 (14.0%)	5,784 (15.2%)	1,404 (3.6%)
Bolted Casks	195 (10.2%)	4,054 (10.6%)	4,624 (12.0%)
Total	1,912	38,117	38,572

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