Materials in Nuclear Waste Disposition

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Commercial nuclear energy has been used for over 6 decades; however, to date, none of the 30+ countries with nuclear power has opened a repository for high-level waste (HLW). All countries with nuclear waste plan to dispose of it in metallic containers located in underground geologically stable repositories. Some countries also have liquid nuclear waste that needs to be reduced and vitrified before disposition. The five articles included in this topic offer a cross section of the importance of alloy selection to handle nuclear waste at the different stages of waste processing and disposal.

INTRODUCTION

Nuclear energy continues to provide worldwide a clean alternative to fossil fuels. Nuclear energy does not release greenhouse gases that may contribute to climate change. Table I shows that currently 31 countries obtain part of their electricity from nuclear power sources.

At present, the United States has the largest number of nuclear reactors (102), which generate approximately 19% of the electricity consumed in the country. France has the largest percentage of nuclear power (75%) that is generated by 58 reactors (Table I).¹ Japan has 50 reactors in operation; however, in 2012, they only generated approximately 2% of the electricity since most of them (~48) were under temporary shutdown after the Fukushima Daiichi tsunami incident on March 11, 2011. Figure 1 shows how many new commercial plants were continuously connected to the grid per year for the last 60 years.¹ The data in Fig. 1 shows that the highest annual number was approximately 30 reactors in the mid-1980s, and since then, it has declined. For the last 10 years, the average number of new reactors connected yearly to the grid was less than five. Figure 1 also shows that 2008 was the only year in which no new reactors were added to the world electrical grid. Figure 2 shows the commercial reactors currently under construction around the globe.¹ It is clear that the largest

amount of new reactors under construction is in Asia including a first reactor in the United Arab Emirates. It appears that most of the reactors are under construction in countries where the mandates for new plants are mostly controlled by the central governments and not by the price of electricity according to demand. Meanwhile, Fig. 2 shows that Western Europe and the Americas seem to be moving away from nuclear energy. After the Fukushima Daiichi incident, some European countries even decided to terminate their nuclear energy programs altogether. Most plants connected to the grid (Table I; Fig. 1) were designed for a lifetime of 40 years. However, many of these plants are requesting and obtaining extensions from regulatory bodies to operate for 60 years and probably for 80 years mainly because no new plants are being constructed.

In the last decade, many reports have correlated climate change to the burning of fossil fuels. The climate change debate has reignited the consideration of alternative sources of energy such as wind, solar, fuel cells, and nuclear power. Of the alternative energies, the amount of electricity produced by nuclear power plants is definitely the largest; however, the percentage varies from country to country, from 75% in France to 19% in the United States to 2% in China.¹ Nuclear power produces the lowest amount of greenhouse gases of any type of energy.^{2,3} However, nuclear power produces nuclear waste, which needs to be isolated from the environment for thousands of years.⁴ All countries currently operating nuclear power reactors agree that the best alternative to deal with nuclear waste is to bury it in geologically stable repositories. The hypothesis is

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Table I. Use of Nuclear Power Worldwide (Source IAEA)¹ (Japan has 50 operating reactors but about 48 are under temporary shutdown. In May 2013, the total worldwide number of reactors in operation was 436)

Country	Percentage of nuclear power of the total in 2012	Number of operating reactors in May 2013	Country	Percentage of nuclear power of the total in 2012	Number of operating reactors in May 2013
France	75	58	Russia	18	33
Slovakia	54	4	Taiwan	18	6
Belgium	51	7	United Kingdom	18	16
Hungary	46	4	Germany	16	9
Ukraine	46	15	Canada	15	19
Sweden	38	10	Argentina	5	2
Slovenia	36	1	Mexico	5	2
Switzerland	36	5	Pakistan	5	3
Czech	35	6	South Africa	5	2
Finland	33	4	India	4	20
Bulgaria	32	2	Netherlands	4	1
Korea	30	23	Brazil	3	2
Armenia	26	1	China	2	18
Spain	21	8	Japan	2	50
Romania	19	2	Iran	1	1
United States	19	102			

that by the very nature of these geological sites, which have not changed for thousands of years, they will contain the waste for long times, limiting their spread, for example, through water flow. Commercial nuclear power has been around for more than 60 years; however, no country is currently operating a nuclear waste repository.⁴ Instead, the spent fuel from commercial power plants is stored in nonpermanent sites, such as water pools or dry storage caskets. Countries that may have some of the most advanced concepts for nuclear waste repositories may include Finland and Sweden. Some of the countries that are currently building most of the new power plants (Fig. 2) do not seem to have current plans for waste repositories.

In a nuclear waste repository, the waste may be confined in metallic containers, which will be in contact with the surrounding environment at the repository. Worldwide, the candidate material for the external layer of the containers include a wide range of alloys, such as carbon steel, copper, titanium, and nickel alloys (Table II).⁴

WORLDWIDE CHARACTERISTICS OF THE PROPOSED REPOSITORIES

More than 30 countries are currently studying the options for disposing of high-level waste (HLW) in deep stable geologic formations, which will be the primary barrier for accomplishing this isolation. All the repository designs also plan to delay the release of radionuclides to the environment by the construction of engineered barrier systems that are planned to be installed to limit water reaching the repository and to restrict radionuclide migration from the waste. In most of the planned repositories, the groundwater associated with the containers should be relatively benign to most materials because of their low ionic strengths, near neutral pH, and in most cases, low concentrations of halide ions.⁵ In most of the repositories (e.g., Finland, Sweden, Canada, and France), the environments will be fully saturated with water (located below the water table) but free of oxygen (anoxic or reducing). For the now-on-hold U.S. repository, the environment was planned to be nonsaturated with an unrestricted ingress of oxygen.⁴ The corrosiveness of the repository waters could increase if significant vaporization occurs due to heating from radioactive decay during the early times of emplacement. Even though the environments *per se* are nonaggressive, corrosion could still be an important factor in the breach of the containers due to the long contact times ($>10^5$ years).⁴

The geologic repositories may not only contain spent fuel from commercial reactors but also legacy waste from the production of weapons. For example, the United States has liquid waste resulting from weapons production that has been stored for decades waiting for conversion into solid form (vitrification) before disposing in a geologic repository. The

Fig. 1. Commercial power plants connected annually to the grid.

second and third articles in this *JOM* topic, by Bruce Wiersma and John Beavers et al., respectively, deal with the handling of liquid nuclear waste at the Savannah River Site (South Carolina) and the Hanford Site (Washington), respectively.

DEGRADATION MODES OF THE ENGINEERING METALLIC MATERIALS

In general, the metallic materials that will enclose the waste may fail by three corrosion mechanisms: (I) general or uniform corrosion, (II) localized corrosion, and (III) environmentally assisted cracking (EAC). Other types of degradation such as microbiologically influenced corrosion (MIC) could be listed as acceleration factors in either uniform corrosion or localized corrosion. King provided a decision tree approach to determine whether MIC would be an important factor in determining the lifetime of the containers.⁶ The modes of corrosion of several types of candidate alloys (carbons steel, copper, stainless steels, nickel alloys, and titanium) are described in the fourth article with precision and detail by Martín Rodríguez.

- (1) General corrosion is the uniform thinning of the containers by interaction with the environment. This is the most predictable type of corrosion because it can easily be incorporated into the design by adding a corrosion allowance for the total design period.
- (2) Localized corrosion is an insidious type of corrosion that develops as discrete sites on the surface while the rest of the container may remain practically unattacked. Localized corrosion may only happen when the rest potential or corrosion potential (E_{corr}) of the container in the repository environment reaches a critical potential (E_{crit})

Fig. 2. New commercial nuclear power plants under construction worldwide.

that is needed for localized corrosion to nucleate and propagate. That is, if $E_{\text{crit}} - E_{\text{corr}} > 0$, localized corrosion will not occur.⁷ It has been shown repeatedly through laboratory testing and modeling that even though localized corrosion may initiate, it may not perforate the container because the attack eventually stifles or stops mainly due to the lack of cathodic support to continue the anodic dissolution.^{8–10}

- (3) EAC could include subgroups such as hydrogen-induced cracking or stress corrosion cracking promoted by sulfide, nitrite, or chloride. Three main factors (e.g., susceptible material, stress, and environment) are needed simultaneously for EAC to occur. If one or more factors are removed from the system, EAC will not occur. For example, if the tensile stresses are removed, cracking will not occur.¹¹

Latest Developments on the U.S. Yucca Mountain Project (YMP)

By 2008, the United States had one of the most advanced studies for a repository worldwide, which was planning to confine its nuclear waste at a remote desert site in Nevada. The container for the waste was going to be a double-walled cylinder having a 2.5-cm-thick layer of Alloy N06022 on the outside and a 5-cm-thick layer of nuclear-grade type 316 stainless steel in the inside.⁴ The U.S. Department of Energy (DOE) supported research, and the development of a Yucca Mountain License Application created an enormous amount of technical and scientific information. After more than two decades of scientific investigations, on June 3, 2008, the DOE submitted to the Nuclear Regulatory Com-

Table II. Metallic materials investigated for waste containers

Metallic material	Explored by country
Carbon steel, low alloy steel, cast iron	USA, France, Japan, Canada, Sweden, Finland, Germany, Argentina, Belgium, Switzerland
Copper, Cu alloys	Canada, Finland, Sweden, Argentina, Japan, USA
Stainless Steel	Belgium, France, USA
Titanium	Canada, Japan, Germany, USA
Nickel Alloys	USA, Germany, France, Argentina

mission (NRC) a License Application to build the repository. The NRC accepted this application on September 8, 2008 (Table III).^{12,13}

In 2010, funding for the YMP was eliminated from the federal budget, and the DOE notified the NRC of its intention to withdraw the Yucca Mountain License Application.¹³ On September 30, 2010, the Office of Civilian Radioactive Waste Management (OCRWM) ceased all its activities. A year later, the NRC Commissioners decided to halt the Yucca Mountain licensing proceedings.¹³ As an alternative to the cancellation of the Yucca Mountain plans, the federal government of the United States named in early 2010 a Blue Ribbon Commission (BRC) on America's Nuclear Future to seek advice on how to deal with the nation's nuclear waste (Table III).¹⁴ The BRC issued its final report and recommendations on January 26, 2012. Some key recommendation points were as follows: (I) the nation should use a new, consent-based approach to siting future nuclear waste management facilities, (II) provide access to the funds nuclear utility rate payers are providing for the purpose of nuclear waste management, (III) develop one or more geologic disposal facilities and other (nonpermanent) storage facilities, and (IV) provide support for continued U.S. innovation in nuclear energy technology and for workforce development, etc.¹⁴ At this moment it is not clear what would be the near-future outcome of the YMP and the BRC Recommendations. Meanwhile, U.S. nuclear plants are planning to store spent fuel not only in pools but also in dry caskets.

Repository Plans Around the World

The United Kingdom has recognized that the geological disposal of the waste in a mined repository is the best available approach.^{15,16} In June 2008, the U.K. government issued the white paper "Managing Radioactive Waste Safely," where a framework for implementing geological disposal is outlined. The location for the repository will be defined through geological screening and community engagement. To complement the permanent repository studies, the U.K. Committee for Radioactive Waste Management has also recommended a robust program of interim storage.¹⁶

The Finnish repository will be located in crystalline bedrock at Olkiluoto Island on the western

coast of Finland.¹⁷ The canister structure consists of a massive cast iron insert covered by a 49-mm-thick copper overpack. Copper has been chosen as the shell material because of its good thermal and mechanical properties and for its resistance to corrosion in reducing environments. Cast iron has been chosen for the insert to provide mechanical strength and radiation shielding and to maintain the fuel assemblies in the required configuration.¹⁸ The repository in Finland should start operations in 2020.¹⁷

Sweden has elected the Forsmark site for its underground repository in the municipality of Östhammar, 500 m below ground in crystalline rock.¹⁹ On March 16, 2011, the SKB applied for the permits needed for the final repository in accordance with the Swedish Act on Nuclear Activities. The waste will be packed in canisters consisting of load-bearing inserts of nodular cast iron surrounded by a 5-cm-thick corrosion barrier of copper. The sealed canister has a total length of 4835 mm and a diameter of 1050 mm.¹⁹ In the repository, the canister will be surrounded by bentonite clay. Both copper and bentonite are naturally occurring materials. The Swedish repository is scheduled to open in 2023, and it is designed to contain the waste for 100000 years.¹⁹

The Japanese Final Disposal Plan calls for a repository that will start operating in the late 2030s.²⁰ It is planned to dispose of the waste in a stable host rock formation more than 300 m underground. The final site for the repository has not been selected yet, but two underground research laboratories have been selected, one 1000 m deep in crystalline rock in the presence of fresh water and the second in sedimentary rock 500 m deep in the presence of saline water.²¹ In the final Japanese repository, the metal containers will be surrounded by bentonite buffer material. The final material for the container has not been selected yet, but it is reported that a thick steel container surrounded by a bentonite buffer overpack would be robust design.²¹

The waste disposal for the French nuclear industry has been outlined in the document Dossier 2005 and calls for the commissioning of a disposal facility by the year 2025.^{22–24} An important concept in the design of the French repository is its reversibility or design evolution at all steps for at least

Table III. Latest timeline of the YMP

Date	Action
June 3, 2008	With two decades of data, DOE submits the License Application to the NRC to build a repository at Yucca Mountain
September 8, 2008	The NRC accepts the License Application
Late 2009	No funding in the FY2010 federal budget for YMP
January 29, 2010	The BRC on America's Nuclear Future was created to provide recommendations for developing a safe, long-term solution to managing the nation's used nuclear fuel and nuclear waste
March 3, 2010	The DOE officially files a motion, with prejudice, through the NRC Atomic Safety and Licensing Board to withdraw the License Application filed in 2008 under the previous administration
September 30, 2010	The DOE OCRWM (in charge of YMP) ceased all its activities
September 12, 2011	The NRC Commissioners decided to halt the Yucca Mountain licensing proceedings
January 26, 2012	The BRC issued its final report and recommendations. One BRC recommendation said that the United States should "[d]evelop one or more geologic disposal facilities and other (non-permanent) storage facilities"
August 13, 2013	The U.S. Court of Appeals for the District of Columbia Circuit said that the NRC should have continued in 2011 the review of the License Application submitted by the DOE

100 years. The cylindrical containers for the HLW (type C in a glass matrix) will be made of standard steel 5 cm thick, 60 cm diameter, and approximately 1.5 m long. It is estimated that this container will remain leak-proof for 4000 years.²² The spent fuel container may have a wall thickness of greater than 10 cm and last 10000 years. Lithuania has recently announced that will it follow the French design for its own repository.

Until recently Germany was considering a salt dome at Gorleben as a site for nuclear waste disposal. In June 2013, Germany's Bundestag decided to search nationwide, via a commission of experts, for a new, more suitable nuclear waste disposal site. In 2014, a federal organism will be created to oversee the waste disposal and a final repository for nuclear waste, which will be selected by 2031. In the aftermath of the Fukushima Daiichi events, Germany is planning to shut down all its commercial reactors by 2022.

Switzerland is studying several sites for waste disposal.²⁵ The geologic site most likely will be Opalinus Clay, and the engineered barriers may include a carbon steel container surrounded by bentonite. Details of the Swiss program are offered in the first article in this topic by Nikitas Diomidis and Lawrence Johnson.

The five articles that follow in this topic contain a small sample on the importance of understanding the behavior of materials (metals) in the entire cycle of nuclear waste management.

- (1) The first article by Nikitas Diomidis and Lawrence Johnson titled "Materials Options and Corrosion-Related Considerations in the Design of Spent Fuel and High-Level Waste Disposal Canisters for a Deep Geological Repository in Opalinus Clay" contains a detailed yet succinct

description of the nuclear waste repository plans by Nagra in Switzerland.²⁵

- (2) The second article titled "The Performance of Underground Radioactive Waste Storage Tanks at the Savannah River Site: A 60-Year Historical Perspective" by Bruce Wiersma describes the behavior of carbon steel tanks as a containment material for liquid waste at the Savannah River Site.
- (3) The third article titled "Corrosion Management of the Hanford High-Level Nuclear Waste Tanks" by John Beavers, Narasi Sridhar, and Kayle D. Boomer describes the behavior of carbon steel tanks as a containment material for liquid waste at the Hanford Site in the state of Washington.
- (4) The fourth article by Martín Rodríguez titled "Anticipated Degradation Modes of Metallic Engineered Barriers for High-Level Nuclear Waste Repositories" is a review on the likely degradation modes of several metals and alloys that are candidates to fabricate waste containers around the globe. Depending on the environment, each alloy or group of alloys is expected to have distinctive degradation modes.
- (5) Finally, the fifth article by Fraser King titled "Predicting the Lifetimes of Nuclear Waste Containers" describes the evolution of the container integrity as a function of time, according to different degradation modes and the special characteristics of each repository.

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