Simulating Water Ingress during Long-Term Storage of Spent Sodium-bonded SMR Fuel

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ABSTRACT

The long-term storage of spent sodium-bonded fuel from Small Modular Reactors (SMRs) presents unique challenges, particularly with respect to water ingress into deep geological repositories (DGRs). This research focuses on the behavior of HT9 cladding, a ferritic-martensitic stainless-steel alloy, when exposed to simulated groundwater conditions in a DGR. Utilizing experimental methods developed in collaboration with CanmetMATERIALS, HT9 coupons are coated with sodium and subjected to controlled tests simulating water ingress. Additionally, a porewater recipe developed by the GeoEngineering Centre of Queen's University and Royal Military College simulates the groundwater chemistry in a DGR. These experiments aim to investigate the chemical reactions and material degradation that occur when groundwater interacts with sodium-bonded fuel cladding. A custom sodium testing apparatus is being constructed to monitor the effects of water ingress under anaerobic conditions. The findings from this study will provide insights into the corrosion mechanisms of sodium-bonded SMR fuel cladding, informing the long-term safety and integrity of nuclear waste storage.

KEYWORDS: sodium-fast reactor, deep geological repository, small modular reactor, spent nuclear fuel, sodium-water interactions

1. INTRODUCTION

The Nuclear Waste Management Organization (NWMO) is a not-for-profit organization formed in 2002 by the nuclear energy producers within Canada. This organization was created in accordance with the Nuclear Fuel Waste Act and aimed to develop an approach for the long-term storage of spent nuclear fuel. NWMO has since established an adaptive phase management plan for spent nuclear fuel, which involves storing the spent fuel within deep geological repositories (DGR). A DGR is a storage facility, 500 meters below ground, with a multi-barrier safety system to safely contain and isolate spent nuclear fuel. The safety system involves placing the spent fuel bundles within used fuel containers (UFC), storing them underground, and packing them with bentonite clay. This system is illustrated in figure 1 [1].

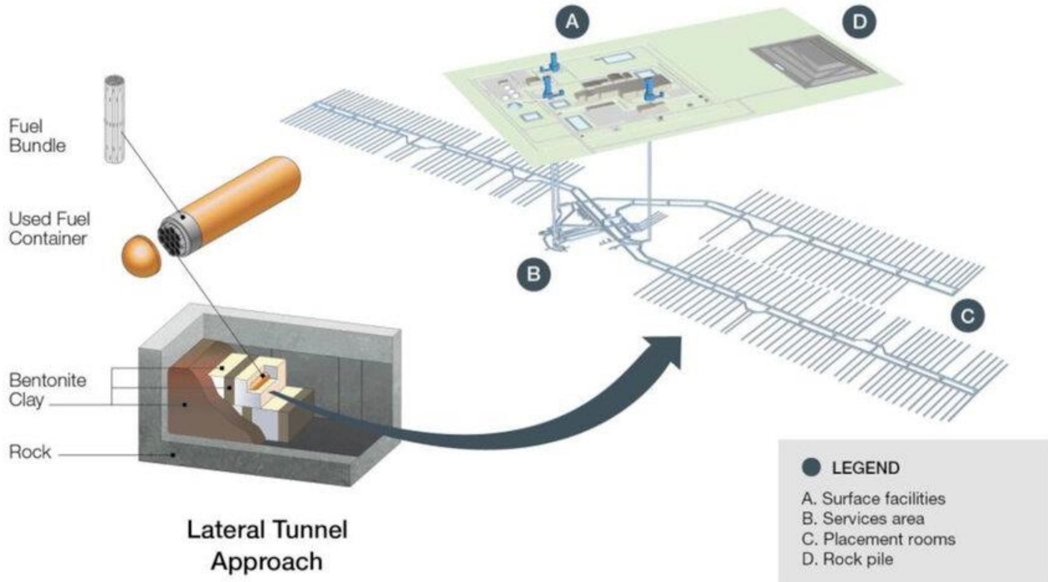

Figure 1. Key Components of a Deep Geological Repository

The current safety assessments of spent nuclear fuel within DGRs are exclusively with Canadian Deuterium Uranium (CANDU) reactors. This study is focused on the safety assessments of Small Modular Reactors, which are slowly being implemented within Canada. Much of the knowledge for sodium fast reactors comes from the Experimental Breeder Reactor II (EBR-II). This reactor, which ran from 1964-1994, was the first experimental reactor that used irradiated fuel, and it also utilized many types of fuel pin configurations. EBR-II is the basis of all experimental experience of sodium fast reactors.

Many SMRs, including ARC-100, use uranium alloy metallic fuel with liquid sodium bonding. The liquid sodium bonds the fuel slug and the cladding, creating a thermally efficient bond, and also allows for swelling. Through fuel irradiation, the sodium both diffuses into the fuel and also displaces in a plug-like form above the fuel slug. Sodium is highly reactive with water and introduces a large risk when considered within the DGRs. The interactions with sodium and water can cause many reaction products, generating hydrogen gas, heat, and many other radioactive contaminants. These contaminants, if in contact with the water, may cause future environmental and safety concerns [2].

The objective of this research is to experimentally determine the reaction products and material effects of the groundwater reactions with sodium-bonded SMR fuel cladding. This research project has finished the literature review and experimental apparatus proposal phase and will be moving on to building and testing.

2. EXPERIMENTAL MATERIALS

This study focuses on the interaction between HT9 fuel cladding and groundwater within a DGR. HT9 is a commonly used cladding material within advanced nuclear systems, due to its irradiation resistance. To simulate the environment within the DGR, a simulated porewater solution is used. The aim is to evaluate the long-term corrosion and degradation of the interaction between the HT9 cladding and simulated groundwater.

2.1. HT9 Cladding and Sodium Coating

HT9 is a ferritic-martensitic stainless-steel alloy widely used as cladding material in advanced reactors. HT9 is so commonly used due to its strong mechanical properties, corrosion resistance, and most importantly, its tolerance to neutron irradiation. Its body-centered cubic (bcc) crystal structure contributes to its lower displacements per atom (dpa), which makes it resistant to damage from high neutron fluxes, a critical factor in sustaining performance under irradiation conditions [3] [4].

The preparation of HT9 coupons follows a systematic process, using roughly the following steps:

1. Melt Sodium at a temperature greater than 100°C in a stainless-steel crucible, within an inert atmosphere glovebox.
2. Dip coat the HT9 coupons in liquid sodium. Weigh the coupons before and after the dip coating to ensure the mass of sodium is measured.
3. Cool the sodium-bonded coupons and store them within sealed containers to ensure contamination is not introduced.
4. Load the coupons into a custom test cell for leak tests and further material evaluation is to be completed.

Figure 2 shows an example of a dip coater, which can be used with the sodium and HT9 coupon.

Figure 2. MTI™ Dip Coater.

Following the leak tests, the last stage of the experiment is simulating the long-term corrosion and material degradation of the fuel cladding by exposing the coupons to alkaline solutions and introducing defects to simulate prolonged exposure in a DGR.

2.2. Simulated Groundwater Conditions

To simulate the groundwater conditions of the DGR, a porewater recipe was developed by the GeoEngineering Centre at Queen's University and Royal Military College. This collaborative effort between institutions was established as a middle ground for the conditions that will likely be found in the Canadian DGR. Table 1 displays the composition of this groundwater recipe [5].

Table 1. Simulated Porewater Recipe

Compounds	Purity	Mass to prepare Brine (g/L)	Cl	Na	Ca	K	Mg	SO_4	H_2O
NaCl	-	152.0	92.2	59.8	-	-	-	-	-
$CaCl_2 \cdot 2H_2O$	100%	79.5	38.3	-	21.7	-	-	-	19.5
KCl	99.8%	96.8	37.1	-	-	40.9	-	-	18.8
$MgCl_2 \cdot 6H_2O$	100%	88.2	30.7	-	-	-	10.5	-	46.9
$MgSO_4 \cdot 7H_2O$	99.4%	1.6	-	-	-	-	0.2	0.6	0.8
Total TDS	-	-	198.4	59.8	21.7	40.9	10.7	0.6	86.0

3. EXPERIMENTAL METHODS

The proposed apparatus of this study is referenced from a sodium leak apparatus and is comprised primarily of an argon-purged vacuum chamber. The sodium-coated HT9 coupons are loaded into the chamber, where dynamic drip tests will be performed to measure changes in hydrogen concentration and heat production. It is expected that the DGRs will be under anaerobic conditions; therefore, the test cell will mimic these conditions. The chamber has eight total components:

- Vacuum Testing Chamber
- Gas Sampling Pump
- Oxygen Analyzer
- Hydrogen Analyzer
- Argon Injection System
- Dripping Lines
- Water Catch Pan
- Weighing Machine

An example of this test apparatus is shown in figure 3 [6] [7].

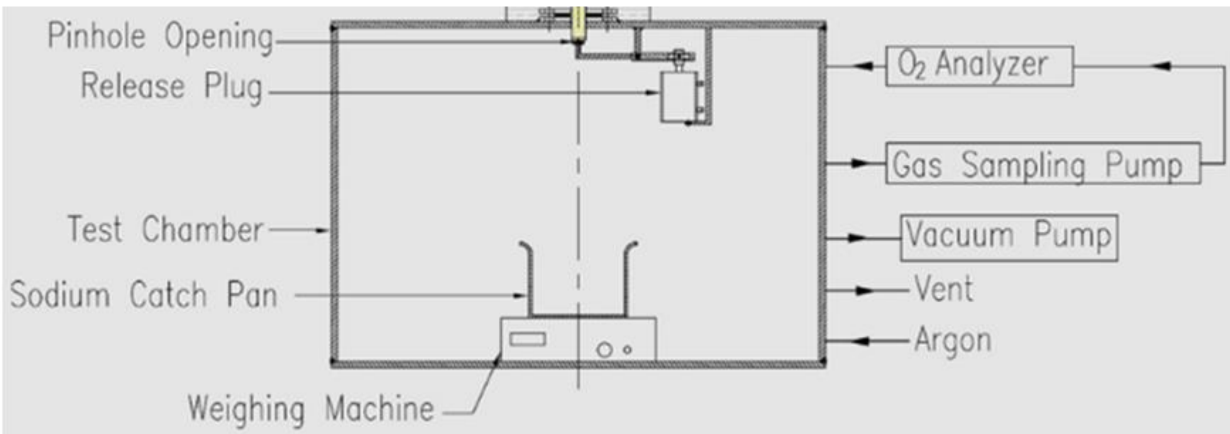

Figure 3. Drip Injection Cell – Example Apparatus

The second apparatus is proposed for further temperature and humidity-dependent experiments. It has the same setup as the first apparatus, with additional temperature and humidity controls. Table 2 outlines the simulated damage on the HT9 coupons, individually testing various damage conditions. Table 3 outlines the testing conditions with the second apparatus [8].

Table 2. Coupon Damage Matrix

Coupon #	Machining Method	Condition
A	Control	Untreated
B	Heat Treatment	Held at 445°C for 100h
C	Mechanical	Thinning Areas by Pressure Punching Mold
D	Electroplating	Zirconium Plating on Mechanically Damaged HT9
E	Corrosion Loop	Under Oxidative Conditions

Table 3. Testing Conditions Summary			
Temp. and RH	**Water Dripping Rate**	**Test Duration**	**Specimens**
80°C / 85% RH	80 mL / day	37 Days	A / B / C
95°C / 75% RH	65 mL / day	67 Days	A / D / E
60°C / 95% RH	80 mL / day	70 Days	A / C / E

4. SODIUM CONTENT IN FUEL

An important aspect of the cladding material is the amount of sodium that is contained within the cladding itself. The corrosion and material degradation are entirely dependent on the amount of sodium on the cladding. The following calculation quantifies the amount of sodium on the cladding, which is directly used in the dip coating procedures. Figure 4 shows the sodium-bonded fuel pin, both before and after reactor irradiation. It is important to note the movement of the sodium as the fuel is irradiated [9].

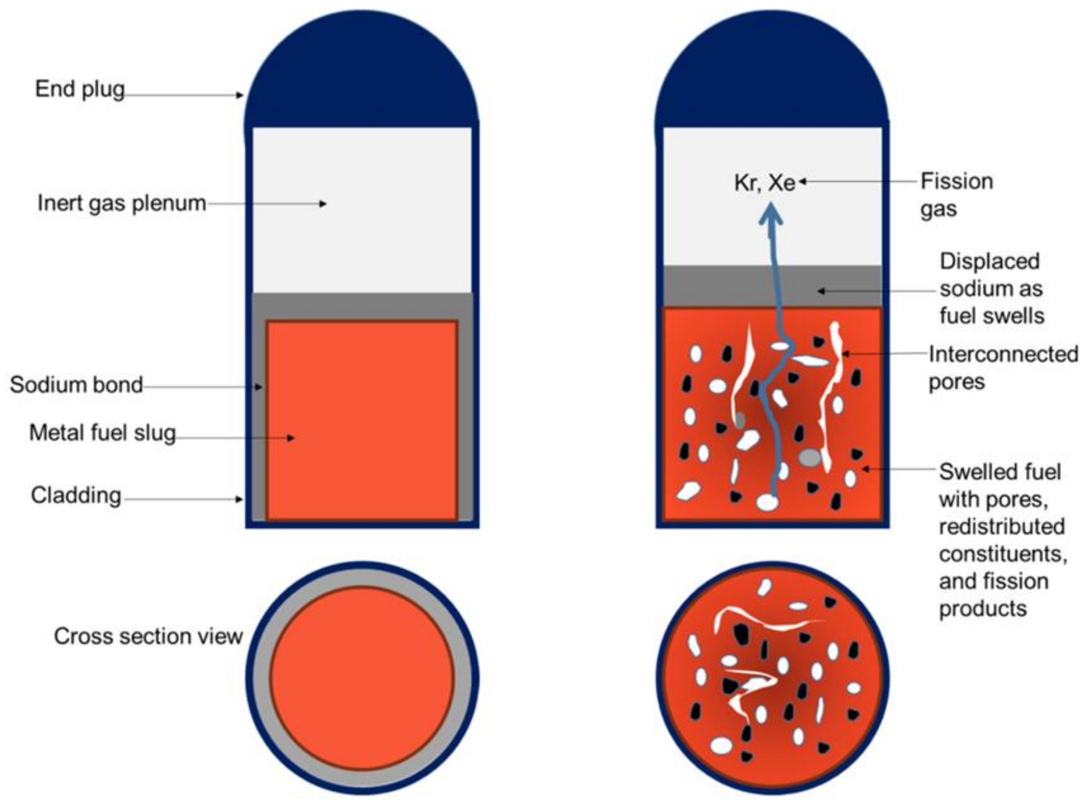

Figure 4. Unirradiated fuel (left) and irradiated (spent) fuel (right)

The variables under analysis are the following:

$$Fuel\ Slug\ Radius\ = r = 0.249\ cm$$
$$Fuel\ Pin\ Radius\ (Inside) = R = 0.315\ cm$$
$$Sodium\ Above\ Fuel\ Slug = h = 2.54\ cm$$
$$Fuel\ Slug\ Length = l = 91.44\ cm$$
$$Fuel\ Pin\ Length = L = 238.1\ cm$$
$$Density\ of\ Sodium\ (25^oC) = \rho_{Na} = 0.968\ g/cm^3$$
$$Density\ of\ Fuel\ (Theoretical) = \rho_F = 15.8\ g/cm^3$$

Assumptions: 35% Fuel Swelling and 57% of Porosity filled by Sodium.

The following characteristics can be calculated and used to determine the total amount of free sodium within the fuel.

Fuel Slug Volume

$$V_F = \pi r^2 l = 17.81\ cm^3$$

Sodium Smear Volume

$$V_S = \pi l(R^2 - r^2) = 10.70\ cm^3$$

Sodium Plenum Volume

$$V_P = \pi R^2 h = 0.792\ cm^3$$

Total Mass of Free Sodium

$$m_{Na,T} = \rho_{Na}(V_P + V_S) = 11.12\ g$$

Swelled Fuel Length

$$L_{SF} = (1.35)l = 123.44\ cm$$

Mass of Logged Sodium

$$m_{Na,L} = (0.57)(0.35)\rho_{Na}\pi R^2 L_{SF} = 7.43\ g$$

Mass of Free Sodium

$$m_{Na,F} = m_{Na,T} - m_{Na,L} = 3.69\ g$$

This calculation gives the quantity of sodium used in the dip coating – 3.69 grams of sodium per HT9 coupon. This is a rough estimate of the irradiation conditions of the fuel within the DGRs.

5. CONCLUSIONS

Collaboration with CanmetMATERIALS is in progress to prepare HT9 coupons and develop the necessary equipment for conducting experiments on these samples. Currently, a custom sodium testing apparatus is being designed and built at CanmetMATERIALS' laboratory. This apparatus will be used to simulate the effects of water ingress on sodium-coated HT9 coupons.

ACKNOWLEDGMENTS

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REFERENCES

1. J. Noronha, “Deep Geological Repository Conceptual Design Report”, 2016.
2. G. L. Hofman et al., “Metallic Fast Reactor Fuels”, 1997.
3. W. J., Carmack, "Temperature and Burnup Correlated FCCI in U-10Zr Metallic Fuel", 2012.
4. Y. Chen, "Irradiation Effects of HT-9 Martensitic Steel", 2013.
5. A. S., Acikel et al., "The impact of multi-component hypersaline wetting on soluble and exchangeable cations and water retention behaviour of MX80 bentonite", 2019.
6. A. Chitella et al., "Experimental investigation on sodium leak behaviour through a pinhole" 2021.
7. C. Morgan, "Phase 2 Initial Borehole Drilling And Testing, South Bruce", 2023.
8. H. Jung et al., "Corrosion Of Alloy 22 And Titanium Alloys Under Seepage Water Dripping Condition", 2011.
9. N. Hall et al., “Storage experience with spent (irradiated) advanced reactor fuel types,” 2019.