

**EFFECTS OF BICARBONATE IONS AND DISSOLVED
HYDROGEN ON DISSOLUTION OF SIMULATED
SPENT NUCLEAR FUEL—PROGRESS REPORT**

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ABSTRACT

The focus of this work was to evaluate the effects of bicarbonate ions and dissolved hydrogen on dissolution of simulated spent nuclear fuel (SNF) in simulated granitic groundwater under oxidizing and reducing conditions. Electrochemical and leaching experiments were conducted using SIMFUEL samples consisting of (i) pure UO_2 , (ii) 35 GW-day/MTU burnup equivalent, and (iii) 60 GW-day/MTU burnup equivalent. Three types of simulated granitic groundwater used in this study were prepared with and without addition of bicarbonate salt. The short-term electrochemical tests were conducted in the granitic groundwater with 0.1 M bicarbonate ions under both oxidizing and reducing conditions. The dissolved oxygen concentration under oxidizing conditions was approximately 8 ppm, and the dissolved hydrogen concentration under reducing conditions was approximately 31 μM . The 2-month-long leaching tests were run in the granitic groundwater with 8.9×10^{-4} M bicarbonate ions and without bicarbonate ions under a high dissolved hydrogen concentration of approximately 3.2 mM.

Electrochemical impedance data for SIMFUEL samples collected in the granitic groundwater with 0.1 M bicarbonate ions were used to compute dissolution rates. The results are compared to previously reported results from experiments that used granitic groundwater with 8.9×10^{-4} M bicarbonate ions and without bicarbonate ions. In comparison with the dissolution rates obtained in the solution with 8.9×10^{-4} M bicarbonate ions, the presence of a high bicarbonate ion concentration of 0.1 M results in increased dissolution rates for all three SIMFUEL samples by a factor of 3 to 8 under oxidizing conditions and a factor of 5 to 16 under reducing conditions. However, under both oxidizing and reducing conditions the dissolution rates generally remained in the same order of magnitude in the solution with 0.1 M bicarbonate ions compared to the solution without bicarbonate ions.

In the leaching tests of the 60 GW-day/MTU SIMFUEL samples in the granitic groundwater under reducing conditions with 3.2 mM dissolved hydrogen, the dissolution rate in the solution with 8.9×10^{-4} M bicarbonate ions is higher than that without bicarbonate ions. The results also showed that the dissolution rates are slightly higher in the solution with 3.2 mM dissolved hydrogen compared to the solution with 1 mM dissolved hydrogen. Nevertheless, complete suppression of SIMFUEL dissolution was not evident in this study under reducing conditions with a high dissolved hydrogen concentration of 3.2 mM.

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QUALITY OF DATA, ANALYSES, AND CODE DEVELOPMENT DATA

DATA, ANALYSES, AND CODES: All CNWRA-generated data contained in this report meet quality assurance requirements described in the CNWRA Quality Assurance Manual. Sources of other data should be consulted for determining the level of quality of those data.

The computer software MATLAB (MATLAB, 2008) was used in the analyses contained in this report. MATLAB is a commercial software controlled under the CNWRA Technical Operating Procedure (TOP)–018, Development and Control of Scientific and Engineering Software. Documentation for experimental data, and dissolution rate calculations can be found in Scientific Notebook 1318 (Pan et al., 2017).

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MATLAB. “MATLAB User’s Guide.” Natick, Massachusetts: The MathWorks, Inc. 2008.

Pan, Y., M. Poerner, and S. Sutton. “Conduct Electrochemical and Leaching Experiments to Determine Dissolution of SIMFUEL under Various Conditions.” Scientific Notebook 1318. San Antonio, Texas: Center for Nuclear Waste Regulatory Analyses. pp. 1-45. 2017.

1 INTRODUCTION

This work was conducted to evaluate the effects of bicarbonate ions and dissolved hydrogen on dissolution of simulated spent nuclear fuel (SNF) in simulated granitic groundwater. The work is a continuation of prior studies reported in Shukla and Ahn (2018) and Shukla et al. (2016).

As discussed in Shukla and Ahn (2018), carbonate (CO_3^{2-}) and bicarbonate (HCO_3^-) ions coexist in solution and their relative concentrations are dependent on pH. Bicarbonate is the predominant species in solutions with pH values below 10. The concentration of bicarbonate ions in a 1 M NaCl solution with a pH of 7 is about three orders of magnitude higher than that of carbonate ions.

Shukla and Ahn (2018) summarized studies on the influence of carbonates (i.e., carbonate and bicarbonate) on SNF dissolution. Literature information suggests that SNF dissolution rates increase by a factor of 10 to 50 in the presence of carbonate and bicarbonate ions under oxidizing conditions (Shoesmith, 2007; Shoesmith, 2000; Oversby and Konsult, 1999). However, carbonate species are not expected to influence SNF dissolution under reducing conditions (Shoesmith, 2007).

Electrochemical experiments were conducted using SIMFUEL in a simulated granitic groundwater with a bicarbonate ion concentration of 8.9×10^{-4} M (Shukla et al., 2016) as well as in a simulated granitic groundwater without bicarbonate ions (Shukla and Ahn, 2018). Under oxidizing conditions, the SIMFUEL dissolution rates were higher in the solution prepared without bicarbonate ions compared to the solution prepared with bicarbonate ions. A similar trend was observed under reducing conditions with dissolved hydrogen concentration of approximately 31 μM . The observed effect of bicarbonate ions on SNF dissolution is contradictory to what is reported in the literature, which is that the presence of bicarbonate ions is expected to increase SNF dissolution under oxidizing conditions.

Shukla et al. (2016) reviewed a large body of literature on the effect of dissolved hydrogen on SNF dissolution. They found that there is no clear consensus on the concentration of dissolved hydrogen needed to completely suppress SNF dissolution under reducing conditions. For example, Wu et al. (2014, Wu et al., 2012) reported that 0.1 to 15 μM of dissolved hydrogen is sufficient to completely suppress SNF dissolution, but Poinssot et al. (2005) and Ferry et al. (2006) concluded that approximately 1 mM of dissolved hydrogen is needed for complete suppression.

The SIMFUEL leaching experiments conducted with a dissolved hydrogen concentration of approximately 1 mM indicate that the presence of dissolved hydrogen decreased SNF dissolution rates in the simulated granitic groundwater with 8.9×10^{-4} M bicarbonate ions (Shukla et al., 2016). The data in Shukla and Ahn (2018) showed that when dissolved hydrogen is approximately 1 mM, the dissolution rates are even lower in the simulated granitic groundwater prepared without bicarbonate ions compared to the solution prepared with bicarbonate ions. Nevertheless, the leaching results suggest that 1 mM of dissolved hydrogen is not sufficient to completely suppress SNF dissolution under reducing conditions.

Considering the lack of clarity on the role of bicarbonate ions and dissolved hydrogen on SNF dissolution rates, additional experiments have been conducted to further evaluate SNF dissolution in simulated granitic groundwater. The scope of the experiments, which are the subject of this report, includes short-term electrochemical tests under both oxidizing and

reducing conditions with a bicarbonate ion concentration of 0.1 M, as well as 2-month-long leaching tests under reducing conditions with a dissolved hydrogen concentration of 3.2 mM.

2 EXPERIMENTAL DETAILS

2.1 SIMFUEL Samples and Test Solution

The experiments were conducted with SIMFUEL specimens in contact with simulated granitic groundwaters at a near-neutral pH (i.e., with calculated pH of 7 and elevated concentrations of Na^+ and Cl^- ions). Dissolution experiments involve a pure UO_2 and two SIMFUEL materials with equivalent burnups of 35 GW-day/MTU (BU35) and 60 GW-day/MTU (BU60). Three simulated granitic groundwater test solutions were used: (1) test solution prepared with 0.1 M bicarbonate ions for electrochemical tests, (2) test solution prepared with 8.9×10^{-4} M bicarbonate ions for one leaching test, and (3) test solution prepared without bicarbonate ions for another leaching test. Details of the original test solution and the SIMFUEL samples used in this work were provided in Shukla et al. (2016).

2.2 Electrochemical Tests

For each SIMFUEL specimen, the electrochemical experiments were conducted under oxidizing and reducing conditions at room temperature:

1. Saturated oxygen condition by bubbling the solution with 1.02 atm [15 psig] compressed air.
2. Saturated hydrogen (and low to negligible oxygen) condition by bubbling the solution with 8.84 atm [130 psig] of a mixture of 4 percent H_2 plus 96 percent N_2 .

Electrochemical impedance spectroscopy was used to measure the SIMFUEL dissolution rates. The dissolved oxygen concentration under Condition 1 was approximately 0.25 mM, and dissolved oxygen was undetectable under Condition 2. The dissolved hydrogen concentration under Condition 2 was estimated to be 31 μM . In each experiment, the working electrode was first polarized to $-1.0 V_{\text{SCE}}$ for 10 minutes. The working electrode was left at the open circuit potential for 16 hours, and electrochemical impedance spectroscopy measurements were subsequently carried out. The electrode potentials were measured during and immediately following the impedance measurements. Details of the experimental setup and procedure used to carry out the experiments were described in Shukla et al. (2016).

2.3 Leaching Tests

In addition to the electrochemical tests, leaching experiments were conducted to determine dissolution rates using leachate concentrations. The objective of these experiments was to measure the dissolution rate under a higher dissolved hydrogen concentration that was near 3.2 mM [compared to 1 mM in the leaching tests conducted in Shukla and Ahn (2018) and Shukla et al. (2016)]. This concentration of dissolved hydrogen can be achieved when the hydrogen fugacity is close to 4.1 atm [60.0 psi]. Two leaching tests were conducted in pipe reactors using BU60 SIMFUEL pellets. The first leaching test was conducted in the simulated granitic groundwater with 8.9×10^{-4} M bicarbonate ions, and the second test in the simulated granitic groundwater without bicarbonate ions. Both reactors were pressurized to approximately 102 atm [1,500 psi] with the 4 percent H_2 plus 96 percent N_2 mixture. Details of the

experimental setup and procedure for the leaching tests were also provided in Shukla et al. (2016).

3 EXPERIMENTAL DATA AND RESULTS

The electrochemical and leaching test data and dissolution rate estimates are presented and described in this section.

3.1 Electrochemical Data

The collected electrochemical data included (1) electrode potential versus time and (2) electrochemical impedance spectroscopy.

3.1.1 Electrode Potential

Figure 1 shows the electrode potential of the UO₂, BU35, and BU60 SIMFUEL electrodes as a function of time in the simulated granitic groundwater containing 0.1 M bicarbonate ions under both oxidizing and reducing conditions at 22 °C [72 °F]. As seen in Figure 1, the electrode potentials reached steady state in about 15–20 hours after initial polarization at $-1.0 V_{SCE}$. The steady-state values of the electrode potential are the corrosion potentials of the electrodes.

The corrosion potentials of the UO₂, BU35, and BU60 electrodes under oxidizing conditions are in the range of -0.1 to $0.1 V_{SCE}$, as seen in Figures 1(a), 1(b), and 1(c), respectively. The corrosion potential values for the three electrodes under oxidizing conditions are in the same range as the values collected for the simulated granitic groundwater without bicarbonate ions under oxidizing conditions (Shukla and Ahn, 2018). This indicates that the corrosion potentials of the three SIMFUEL electrodes under oxidizing conditions were not affected when a high concentration of 0.1 M bicarbonate ions was added to the simulated granitic groundwater.

The corrosion potentials under reducing conditions for the UO₂ electrode are in the range of -0.4 to $-0.5 V_{SCE}$ in Figure 1(a). These values are close to the data collected for the simulated granitic groundwater prepared without bicarbonate ions (Shukla and Ahn, 2018). The same observation was made for the BU35 and BU60 electrodes. The corrosion potentials for the BU35 are in the range of -0.4 to $-0.7 V_{SCE}$ [Figure 1(b)] and the corrosion potentials for the BU60 are in the range of -0.5 to $-0.7 V_{SCE}$ [Figure 1(c)]. The values of the corrosion potentials overlap with data collected in the simulated granitic groundwater without bicarbonate ions. This indicates that the addition of 0.1 M bicarbonate ions does not affect the corrosion potential of BU35 and BU60 electrodes under reducing conditions.

Table 1 lists the dissolved oxygen concentrations of the test solutions measured immediately after the electrochemical tests. These measurements were conducted while the corrosion cell was inside a glove box. An oxygen-free environment was maintained inside the glove box under both conditions by flowing nitrogen gas through the glove box chamber. The measured oxygen concentrations are close to 8 ppm for Condition 1, under which air at 1.02 atm [15 psig] was bubbled through the test solution. This result was anticipated because the solution is expected to be saturated with oxygen under Condition 1. The measured oxygen concentrations are below the detection limit (<0.005 ppm) of the oxygen meter for Condition 2, under which the 4 percent H₂ plus 96 percent N₂ mixture was bubbled through the test solution. This was anticipated, because the nitrogen in the gas mixture is expected to purge the oxygen in the solution.

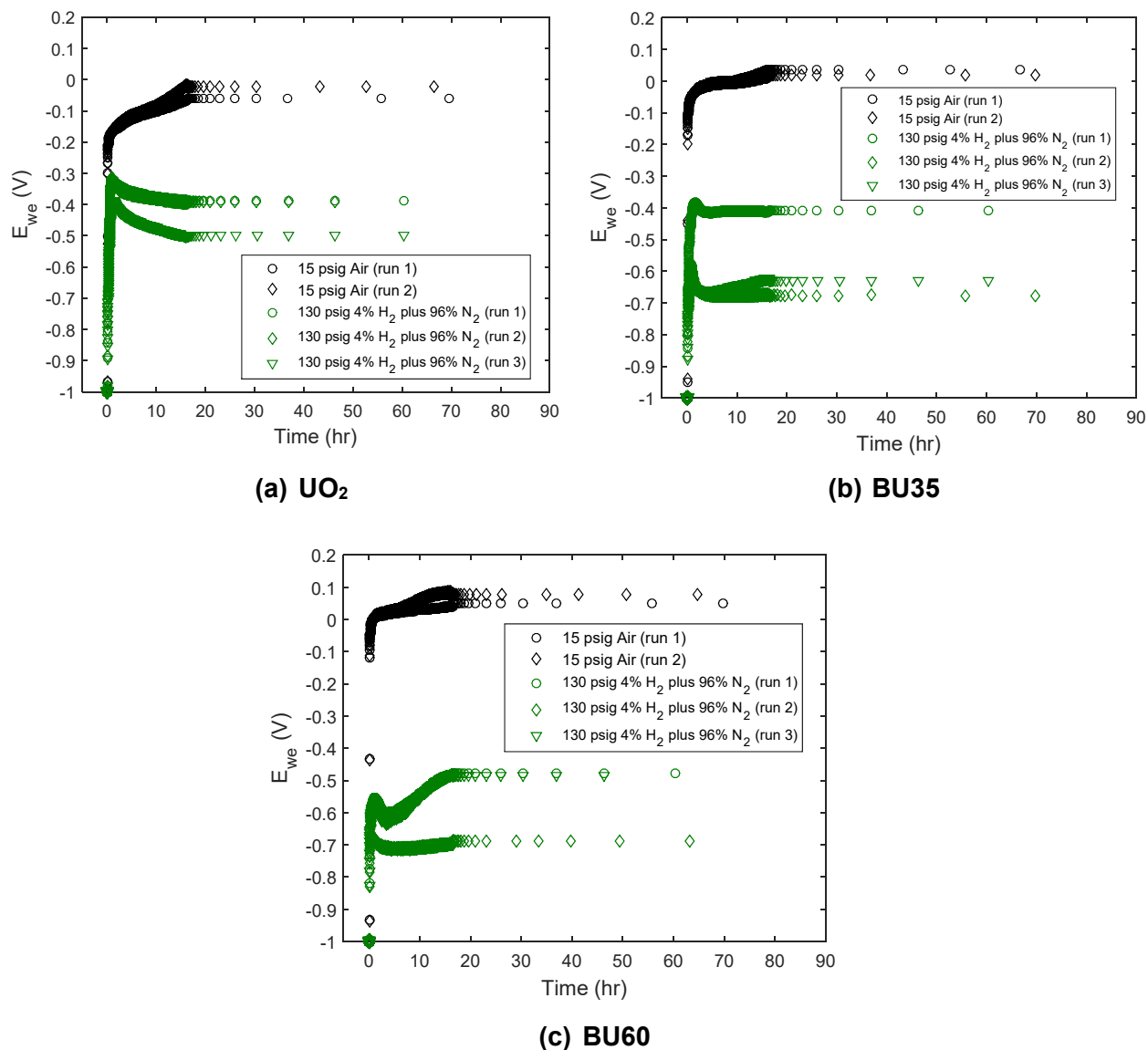


Figure 1. Working electrode potential (E_{we}) with respect to saturated calomel electrode versus time for (a) UO₂, (b) BU35, and (c) BU60 electrodes collected in simulated granitic groundwater prepared with 0.1 M bicarbonate ions at 22 °C [72 °F] under the two conditions

Table 1. Oxygen concentrations in the test solutions immediately after completion of the electrochemical experiments			
Test Condition	Oxygen Concentration (mg/L)		
	UO ₂	35 GW-day/MTU burnup	60 GW-day/MTU burnup
1. 1.02 atmg [15 psig] air	8.02 7.98	8.26 7.93	8.79 9.06
2. 8.84 atmg [130 psig] of 4 percent H ₂ plus 96 percent N ₂	<0.005*	<0.005*	<0.005*

*Detection limit of the instrument

3.1.2 Electrochemical Impedance Spectroscopy

Figure 2 shows the Bode and phase angle impedance spectra for the UO_2 SIMFUEL electrode under the two test conditions in the simulated granitic groundwater. The Bode and phase angle impedance spectra for BU35 and BU60 SIMFUEL electrodes are presented in Figure 3.

As reported in Shukla and Ahn (2018) and Shukla et al. (2016), two time constants are observed in the impedance spectra of each electrode. The first time constant that appears in the high frequency range represents the ohmic resistance due mainly to the intrinsic electrical resistance of the SIMFUEL pellet and resistance of the electrolyte solution to the charge flow. The second time constant that appears in the low frequency range is associated with the polarization resistance at the SIMFUEL electrode surface. For the UO_2 electrode, the first time constant is visible in the frequency range of 10 Hz to higher frequencies, and the second time constant is apparent in the frequency range of 10 Hz to lower frequencies [Figure 2(b)]. Similarly, for the BU35 and BU60 electrodes, the first time constant is visible in the frequency range of 10^4 Hz to higher frequencies, and the second time constant is apparent in the frequency range of 10^4 Hz to lower frequencies [Figures 3(b) and 3(d)].

As seen in Figures 2(a), 3(a), and 3(c), no noticeable change was observed in the impedance spectra collected in the simulated granitic groundwater containing 0.1 M bicarbonate ions compared to the spectra collected in the solution without bicarbonate ions in Figures 3-2 and 3-3 of Shukla and Ahn (2018).

3.2 Dissolution Rate Estimates

Posttest analyses included the collection of (1) electrochemical impedance spectroscopy data and (2) leaching test data. These data were analyzed to estimate the SIMFUEL dissolution rates, as described in the following sections.

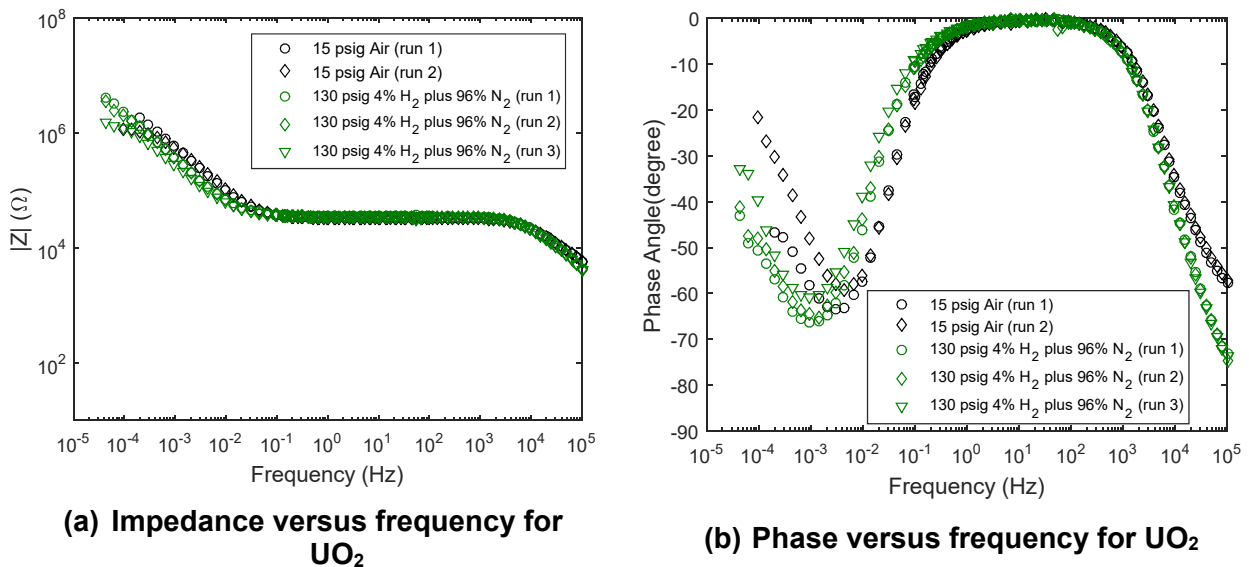
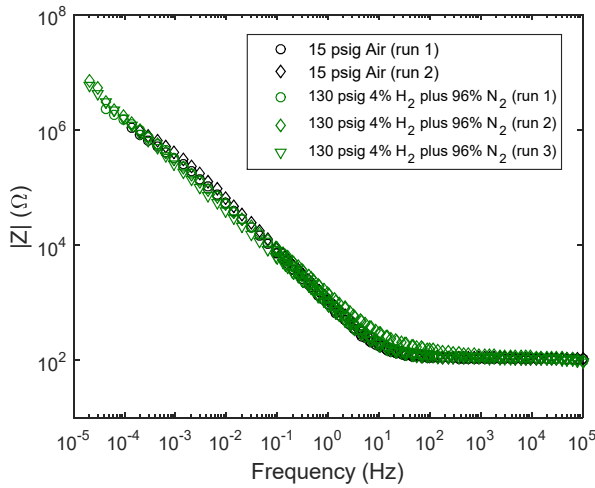
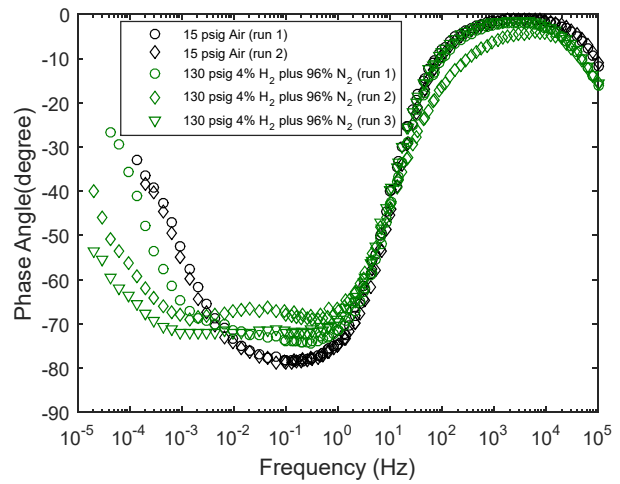


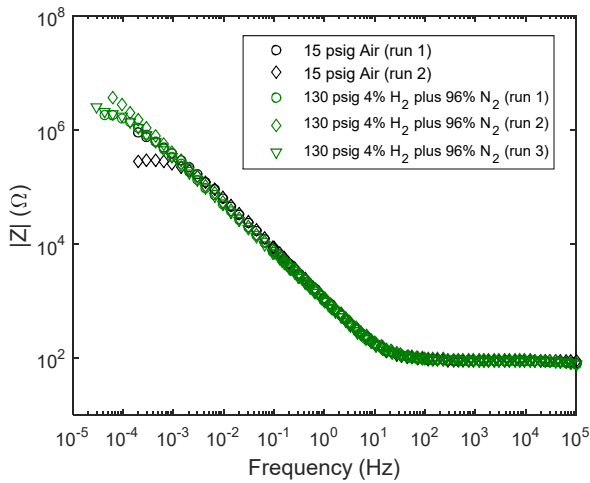
Figure 2. Electrochemical impedance data for UO_2 electrode collected in simulated granitic groundwater prepared with 0.1 M bicarbonate ions at 22 °C [72 °F] under the two conditions



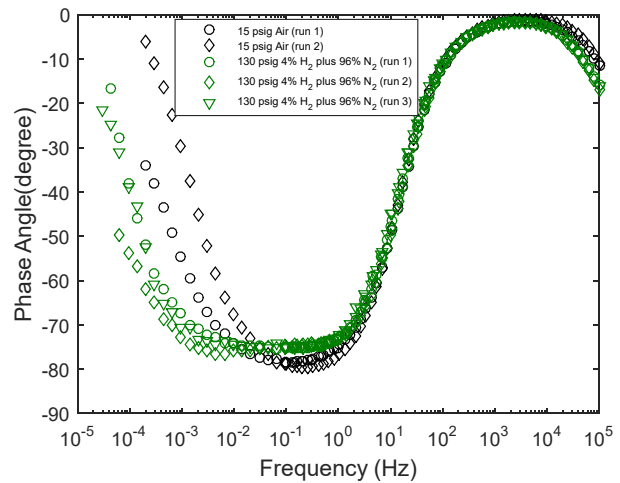
(a) Impedance versus frequency for BU35



(b) Phase versus frequency for BU35



(c) Impedance versus frequency for BU60



(d) Phase versus frequency for BU60

Figure 3. Electrochemical impedance data for BU35 and BU60 electrodes collected in simulated granitic groundwater prepared with 0.1 M bicarbonate ions at 22 °C [72 °F] under the two conditions

3.2.1 Electrochemical Impedance Spectroscopy

The dissolution rates of the SIMFUEL specimens in the simulated granitic groundwater containing 0.1 M bicarbonate ions under the two test conditions were estimated by fitting the impedance data [Figures 2(a), 3(a), and 3(c)] to an electrical circuit model to obtain the polarization resistance (R_p) associated with the second time constant. Description of the circuit model and associated parameters is provided in Shukla et al. (2016). The values of the polarization resistance are listed in Table 2. The polarization resistance was normalized with respect to the surface area of the electrode by multiplying the polarization resistance value by the electrode surface area (0.54 cm^2). Normalized polarization resistance values were used to obtain the corrosion current density (i_{corr}) using the Stern-Geary equation (1957):

Test Condition	Polarization Resistance (Ω)		
	UO ₂	35 GW-day/MTU	60 GW-day/MTU
1. 1.02 atmg [15 psig] air	1.67 × 10 ⁶ (run 1) 1.00 × 10 ⁶ (run 2)	1.51 × 10 ⁶ (run 1) 1.31 × 10 ⁶ (run 2)	1.14 × 10 ⁶ (run 1) 3.20 × 10 ⁵ (run 2)
2. 8.84 atmg [130 psig] of 4 percent H ₂ plus 96 percent N ₂	4.40 × 10 ⁶ (run 1) 3.10 × 10 ⁶ (run 2) 1.56 × 10 ⁶ (run 3)	2.35 × 10 ⁶ (run 1) 9.30 × 10 ⁶ (run 2) 9.19 × 10 ⁶ (run 3)	2.31 × 10 ⁶ (run 1) 6.62 × 10 ⁶ (run 2) 2.73 × 10 ⁶ (run 3)

$$i_{corr} = B / R_{pa} \quad (1)$$

where

- i_{corr} — corrosion current density [A/cm²]
- R_{pa} — normalized polarization resistance [Ω -cm²]
- B — composite Tafel parameter [V]

The value of the composite Tafel parameter B was selected to be 25 mV based on literature data (Grambow et al., 2000; Carbol et al., 2005). The dissolution rate was calculated from the polarization resistance using Faraday's law according to Eq. (2)

$$\text{Dissolution Rate } (D) = K_2 \times i_{corr} \times EW \quad (2)$$

where

- K_2 — constant [8.95×10^6 mg-cm²/A/m²/ day]
- EW — equivalent weight for UO₂ = 33.75 assuming +6 and -2 valences for U and O, respectively

Table 3 lists the estimated dissolution rates of the three SIMFUEL electrodes under the two test conditions. The dissolution rates under oxidizing conditions are in the range of 8.36 to 43.6 mg/m²/day for the three electrodes. The dissolution rates under reducing conditions are in

Test Condition	Dissolution Rate (mg/m ² /day)		
	UO ₂	35 GW-day/MTU	60 GW-day/MTU
1. 1.02 atmg [15 psig] air	8.36 (run 1) 13.9 (run 2) Average=11.1	9.26 (run 1) 10.7 (run 2) Average=9.98	12.2 (run 1) 43.6 (run 2) Average=27.9
2. 8.84 atmg [130 psig] of 4 percent H ₂ plus 96 percent N ₂	3.14 (run 1) 4.50 (run 2) 8.95 (run 3) Average=5.53	5.94 (run 1) 1.50 (run 2) 1.52 (run 3) Average=2.99	6.04 (run 1) 2.11 (run 2) 5.11 (run 3) Average=4.42

the range of 1.50 to 8.95 mg/m²/day. For all three SIMFUEL electrodes, the average dissolution rates under oxidizing conditions are generally higher compared to under reducing conditions.

3.2.2 Leaching Tests

The leaching of the BU60 SIMFUEL pellets was run for 61 days in the simulated granitic groundwater with 8.9×10^{-4} M bicarbonate ions and without bicarbonate ions. Fresh test solution was used for each test. The test solutions were collected after completion of the experiments and analyzed using inductively coupled plasma-mass spectrometry. Uranium concentration data obtained from analysis of the leachate are listed in Table 4. The data were used to estimate the dissolution rate according to the following equation

$$\text{Dissolution Rate } (D) = 2.5 \times 10^{-2} C_U / (S_{\text{pellet}} \times t_{\text{exposure_days}} \times mfu) \quad (3)$$

where

- C_U — uranium concentration [mg/L]
- S_{pellet} — surface area of the pellet [m²]
- $t_{\text{exposure_days}}$ — exposure time [days]
- mfu — mass fraction of U in the pellet [0.9406 for the 60 GW-day/MTU sample]

The numerical factor on the right hand side of Eq. (3) accounts for the 25 mL of the simulated granitic groundwaters used in each experiment. The estimated surface area of the pellets tested in simulated granitic groundwater with and without bicarbonate ions is approximately 3.36×10^{-4} m² [3.62×10^{-3} ft²] and 3.13×10^{-4} m² [3.37×10^{-3} ft²], respectively. The dissolution rates, estimated using the surface area, uranium leachate concentration, and exposure time, are listed in Table 4.

Both the leachate U concentration and the dissolution rate for the test in the simulated granitic groundwater with 8.9×10^{-4} M bicarbonate ions are higher compared to the test in the solution without bicarbonate ions.

Table 4. Uranium concentrations obtained from chemical analyses of the leachate collected after leaching tests and estimated dissolution rates from the uranium concentrations		
Parameter	60 GW-day/MTU sample in simulated granitic groundwater (prepared without bicarbonate ions)	60 GW-day/MTU sample in simulated granitic groundwater (prepared with 8.9×10^{-4} M bicarbonate ions)
Uranium concentration in leachate (mg/L)	2.78×10^{-2}	6.18×10^{-2}
Dissolution rate (mg/m ² /day)	3.78×10^{-2}	8.02×10^{-2}

4 SUMMARY AND DISCUSSION

Dissolution of SIMFUEL samples in contact with simulated granitic groundwater was measured using both electrochemical and leaching methods. The short-term electrochemical tests were conducted in the simulated granitic groundwater with 0.1 M bicarbonate ions under oxidizing and reducing conditions. The dissolved oxygen concentration under oxidizing conditions was approximately 8 ppm, and the dissolved hydrogen concentration under reducing conditions was approximately 31 μM . The 2-month-long leaching tests were run in the simulated granitic groundwater with 8.9×10^{-4} M bicarbonate ions and without bicarbonate ions under a dissolved hydrogen concentration of 3.2 mM.

The calculated SIMFUEL dissolution rates obtained using the simulated granitic groundwater prepared with 0.1 M bicarbonate ions (this study), as well as with the simulated granitic groundwater prepared with 8.9×10^{-4} M bicarbonate ions (Shukla et al., 2016) and without bicarbonate ions (Shukla and Ahn, 2018), are provided in Table 5. An average value and values obtained from multiple runs for each sample under various conditions are reported.

Table 5. Summary of dissolution rate (mg/m²/day) data obtained in simulated granitic groundwaters with and without bicarbonate ions			
Parameter (condition and SIMFUEL sample)	In simulated granitic groundwater (prepared without bicarbonate ions)*	In simulated granitic groundwater (prepared with 8.9×10^{-4} M bicarbonate ions)*	In simulated granitic groundwater (prepared with 0.1 M bicarbonate ions)*
Oxidizing Condition			
UO ₂	15.2 (19.1, 14.4, 12.1)	1.47	11.1 (8.36, 13.9)
35 GW-day/MTU	12.88 (21.1, 4.66)	3.35 (3.62, 3.08)	9.98 (9.26, 10.7)
60 GW-day/MTU	7.97 (11.0, 4.94)	6.20	27.9 (12.2, 43.6)
Reducing Condition (31 μM dissolved hydrogen)			
UO ₂	17.9 (14.9, 20.9)	0.48 (0.42, 0.66, 0.36)	5.53 (3.14, 4.50, 8.95)
35 GW-day/MTU	1.82 (3.22, 0.96, 1.27)	0.54 (0.84, 0.23)	2.99 (5.94, 1.50, 1.52)
60 GW-day/MTU	1.24 (1.15, 1.07, 1.50)	0.28 (0.58, 0.17, 0.23, 0.22)	4.42 (6.04, 2.11, 5.11)
Reducing Condition (1 mM dissolved hydrogen)			
35 GW-day/MTU	1.70×10^{-2}	2.40×10^{-2}	—
60 GW-day/MTU	2.50×10^{-2}	6.10×10^{-2}	—
Reducing Condition (3.2 mM dissolved hydrogen)			
60 GW-day/MTU	3.78×10^{-2}	8.02×10^{-2}	—
*Whenever available, the average value from multiple runs is used. In addition, specific values for multiple runs are provided directly below the average values.			

A comparison of the average SIMFUEL dissolution rates from the electrochemical experiments conducted in the solutions prepared with 8.9×10^{-4} M and 0.1 M bicarbonate ions showed that under oxidizing conditions, the presence of a high bicarbonate ion concentration of 0.1 M results in increased dissolution rates by a factor of 3 to 8. The observed effect of bicarbonate ions is consistent with literature information. A similar trend was observed for SIMFUEL dissolution under reducing conditions; that is, the dissolution rates in the solution with 0.1 M bicarbonate ions are about 5 to 16 times higher compared to those with 8.9×10^{-4} M bicarbonate ions. In addition, the dissolution rates for the three SIMFUEL samples under both oxidizing and reducing conditions generally remained in the same order of magnitude in the solution prepared with 0.1 M bicarbonate ions compared to the solution prepared without bicarbonate ions. The results are contrary to indications from literature data that the presence of bicarbonate ions is not expected to influence SNF dissolution under reducing conditions.

The leaching experiments of BU60 SIMFUEL samples in the simulated granitic groundwater under reducing conditions with 3.2 mM dissolved hydrogen showed that the dissolution rate in the solution with 8.9×10^{-4} M bicarbonate ions is higher than that without bicarbonate ions—similar to what was seen in the leaching experiments performed in the solution with 1 mM dissolved hydrogen. The results also showed that the dissolution rates are slightly higher in the solution with 3.2 mM dissolved hydrogen compared to the solution with 1 mM dissolved hydrogen. Nevertheless, complete suppression of SNF dissolution was not evident in this study with a high dissolved hydrogen concentration of 3.2 mM.

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