H₂ Generation by the ¹⁰B(n,α)⁷Li Reaction in High Temperature Water

Steven J. Guerin^{a,b}, Mohamad I. Al-Sheikhly^a, Alan K. Thompson^c, Cameron S. Goodwin^d, Sangho Nam^d, David M. Bartels^{b^{*}}

^a Dept. of Materials Science and Engineering, Univ. of Maryland, College Park, MD 20742, United States

^b Radiation Laboratory, Univ. of Notre Dame, Notre Dame, IN 46556, United States
^c National Institute of Standards and Technology, Gaithersburg, MD 20899, United States
^d Rhode Island Nuclear Science Center, 16 Reactor Rd., Narragansett, RI 02882, United States

KEYWORDS water radiolysis, ¹⁰B, neutron capture

Abstract H₂ produced in water from the ¹⁰B(n, α)⁷Li fission reaction has been measured up to 300 °C. Thermal energy neutrons from the Rhode Island Nuclear Science Center's 2 MW reactor interact with boric acid-containing water in temperature-controlled highpressure cells made from tubing of either titanium or zirconium alloy. After exposure for a minimum of one hour, the solution sample is extracted and sparged with argon. The H₂ entrained by the sparging gas is sampled with a small mass spectrometer. A small amount of sodium is included in the boric acid solution so that after sparging, samples can be collected for ²⁴Na activation measurements in a gamma spectrometer to determine the neutron exposure and thus the total energy deposited in solution. The G-value (µmol/J) for H₂ production is obtained for water at a pressure of 25 MPa, over a temperature range from 20 °C to 300 °C. The weak temperature dependence of this yield between 150 °C and 200 °C demonstrates that the bimolecular reaction of pairs of e_{aq}^- is a very minor source of H₂ in high LET tracks.

*Corresponding Author. Email: David.M.Bartels.5@nd.edu

1 Introduction

¹⁰B is injected as boric acid into commercial Pressurized Water Reactor (PWR) primary coolant as a soluble (and thus homogeneously dispersed) chemical neutron "shim" due to its relatively large thermal neutron capture cross section of $7.63 \times 10^{-26} m^2$ (763 barn) in order to compensate for fuel burnup over the course of a fuel cycle. The ${}^{10}B(n,\alpha)^7Li$ fission reaction typically produces a ${}^{4}\text{He}^{2+}$ ion with kinetic energy 1.473 MeV and ${}^{7}\text{Li}^{1+}$ ion with kinetic energy 0.841 MeV (with emission of a 0.479 MeV gamma ray), while there is also a 6 % branching ratio for the reaction to instead produce a 1.778 MeV ⁴He²⁺ ion and a 1.015 MeV ⁷Li¹⁺ ion with no associated gamma (Auden, et al., 2019). These ions are characterized as high Linear Energy Transfer (LET) particles and quickly lose their energy by ionizing the aqueous medium, producing H₂, H₂O₂, 'OH radicals, 'H atoms, and e_{aq}^{-} in dense tracks (Mozumder, 1999). This radiation can account for up to 33 % of the total dose to the primary coolant in the reactor core at the beginning of a fuel cycle when the boric acid is kept at its highest concentration (Christensen, 1995). The stable "escape" products from the high LET tracks are mainly H₂ and H₂O₂ (Laverne, 2000; LaVerne, 2004), in nearly equal amounts. Given the importance of this source for corrosive H₂O₂ (Macdonald, 1992; Lin, 2000; Raiman, et al., 2017; Bartels, et al., 2013), the product yields of this event should be accurately included in models of the cooling water radiation chemistry (Elliot & Bartels, 2009). It is surprising that there exist no measurements of any product yields for this radiolysis event in high temperature water, and almost no

measurements at room temperature (Barr & Schuler, 1959; Yokohata & Tsuda, 1974; Dietz, et al., 2021) (Kusumoto, et al., 2020) (Kusumoto & Ogawara, 2019). Estimating the total energy deposited into water from the ${}^{10}B(n,\alpha)^7Li$ reaction is straightforward when given the nuclear cross-sections of boron (Carlson, 2011; Carlson, et al., 2018; Brown, et al., 2018), the neutron flux density, and the boron concentration in the water. This paper presents new measurements of the H₂ yield from the ${}^{10}B(n,\alpha)^7Li$ reaction, at a pressure of 25 MPa and over a temperature range from 20 °C to 300 °C.

2 Experimental Method

The experiment is comprised of two essential measurements: the amount of dissolved molecular hydrogen that was produced from the high LET fission products' water radiolysis, and the amount of ²⁴Na activation in the same volume which is proportional to the thermal neutron exposure of the ¹⁰B. The hydrogen produced can then be normalized to the number of ¹⁰B capture events to deduce a radiation chemical yield, or G-value.

2.1 RINSC Facility

Neutron irradiations were carried out in the thermal neutron column of a 2 MW open pool light water research reactor with a low enriched uranium core, located at the Rhode Island Nuclear Science Center (RINSC) at the Narragansett Bay Campus of the University of Rhode Island. This facility has been described elsewhere (Crow, et al., 1995; Tehan, 2000; RIAEC, January 2017). Neutrons travel through a 2 meter deep stack of graphite blocks to achieve a thermalized spectrum at the end of the column (Mitsui & Siguyama, 1973). The graphite blocks are arranged to form a shallow cavity on the outer face of the stack, where a sample can be placed and irradiated via a diffuse thermal neutron "glow" coming uniformly from the five surrounding faces of graphite. A high thermal neutron flux density is desirable to obtain an acceptable signal-to-noise ratio for the relatively low concentrations of H₂ expected. The neutron fluence was characterized prior to the experiment by using the cadmium-ratio method (Sekine & Baba, 1980). Gold foils both with and without a cadmium cover were placed at the desired experimental location within the thermal column and irradiated. From the analysis of their activation we can expect a flux density for thermal neutrons of 2.4×10^8 s⁻¹ cm⁻². Low gamma background is necessary to isolate the boron fission H₂ from gamma H₂. Using radiochromic film from Far West, Inc. (Butson & Niroomand-Rad, 2017), the gamma background dose rate is estimated as 1.5Gy/hr to 2.0 Gy/hr at the sample location.

The RINSC reactor facility includes two compensated ion chamber detectors located within the pool near the core to monitor the reactivity level in real time. A reading is logged every 5 seconds as a percentage of the maximum 2 MW power output. A plot demonstrating the reactor stability during a daily run is shown in Figure 1, where three sample irradiation periods have been indicated. The three averaged neutron flux densities (the calculation of these values and their uncertainties will be discussed further on) are plotted to show their relative deviation compared with the behavior of the power trend readings during their associated irradiation times.



Figure 1. Plot of the data from power trend readings logged by the compensated ion chamber detectors. The points associated with the first (blue), second (yellow), and third (green) experimental irradiations are distinguished from the readings during reactor power-up and shutdown (red). Also shown are the neutron flux densities calculated by sodium activation in each sample (black).

2.2 Irradiation Cell

Two irradiation cells were constructed for high temperature and pressure, consisting of three tubing sections, each 1.52 m long with a 6.35 mm outer diameter and 4.45 mm inner diameter, plumbed together with 316 stainless steel Swagelok union fittings. The tubing was bent into a rectangular coil ca. 15 cm across to enhance the coupling between the liquid target and the neutron field, and still allow fast quantitative removal of the irradiated liquid by utilizing a flow-through geometry. Total liquid volume of the flow cell is 75 ± 2 mL. The two cells differ only in

the material selected for the main three sections of tubing, with the first cell made of Zircadyne® 702 alloy and the second made of Commercially Pure Grade 2 Titanium. The zirconium alloy is expected to undergo less neutron activation than the titanium tubing, which will introduce error in the G-value of H₂ from delayed gamma fluorescence; titanium is expected to be less likely to corrode at temperatures above 200 °C, where the metal oxidation reaction would also interfere as a source of H₂ overproduction.

The cell tubing is wrapped with high temperature heating tape, along with a layer of aluminum foil and a layer of fused silica batting to insulate the cell while heating up to a maximum of 350 °C. A main thermocouple is in contact with the tubing metal to monitor the liquid for temperature control over the full duration of all irradiations, and a secondary thermocouple monitors the temperature of the air contained within the cell insulation layers to verify temperature uniformity. All other tubing used in the flow system was composed of 316 stainless steel. This full experimental cell "unit" is contained within a lidded tray made of aluminum sheet metal to simplify installation into the cavity on the front face of the graphite in the thermal column.

Unfortunately the thermocouple wire extensions used for the cell thermocouples were hooked up backwards until almost the end of these experiments. The four additional dissimilar metal-metal junction voltages in series were paired so as to nearly subtract out of the electrical voltage measurement, such that the problem was not easily recognized. In retrospect, we believe the temperature readout on any given day may have been uncertain by ± 5 °C. Fortunately the G(H₂) proves to have very weak temperature dependence so that this problem was not critical.

6

2.3 Solutions

The standard experimental borated solution is prepared by mixing 36.68 g of boric acid (Sigma, BioUltra grade, ≥99.5 % trace metal basis), 0.0292 g of sodium carbonate (Fisher, ACS grade, \geq 99.5 % trace metal basis), and 2 L of \geq 10 M Ω -cm deionized water filtered to \leq 100 ppb TOC. This yields a solution of 0.3 M of natural boron (pH 5 at room temperature), with 0.265 mM of ²³Na ions. Solution volumes are deaerated with a gas mixture of (2.50 ± 0.05) % N₂O in argon, obtained from Airgas Inc. Based on its solubility, this should give a 0.57 mM concentration of N₂O to scavenge any aqueous electrons and prevent recombination reactions which might produce additional H₂. The 'OH radicals are expected to react with the CO_3^{-} ions. Alternative experimental solution mixtures substituted 0.0243 g of sodium nitrite (Sigma-Aldrich , >99 %) instead of sodium carbonate (to better scavenge 'OH and especially 'H atoms that manage to escape the tracks), and some include 0.156 g of enriched lithium-7 hydroxide monohydrate (Aldrich, 99.95 % trace metal basis) to ascertain any significant effect from the pH level at room temperature and to more accurately resemble the chemistry (near pH 7) in a PWR at temperatures >200 °C. No effect on H₂ production was noted upon changing these scavengers for the minor radiolysis species, presumably because the gamma background is small, and there is no effect on the radiolysis kinetics in high-LET tracks.

Isotopic ratio analysis of ${}^{10}B/{}^{11}B$ was measured by mass spectroscopy, confirming the ratio in our boric acid sample is the natural fractional abundance of 0.199 ± 0.007 listed in many sources (NIST, 2022). The precise Na⁺ density of stock Na₂CO₃ and NaNO₂ solutions were calibrated relative to a commercial (1000 ± 3) µg cm⁻³ Na⁺ density standard using ICP-OE.

2.4 Apparatus

A schematic of the apparatus used to irradiate solution, collect it, and test it for hydrogen is shown in Figure 2. The irradiation cell is located within the thermal column enclosure, and all other components of the full experimental apparatus are located ca. 3 meters away within a heated commercially available greenhouse enclosure to mitigate the large temperature fluctuations which are common within the RINSC facility's reactor vault. The water and chemicals are added to a glass reservoir and mixed by a magnetic stir bar for at least 30 minutes. The solution is then drawn from its reservoir into a Teledyne ISCO 260D syringe pump, which can then force solution to flow out either of its two outlets. One outlet is routed to bypass the rest of the system and leads directly to the glass sparging apparatus to avoid unwanted irradiation of select solution volumes while the reactor is powered up. The other outlet is sent towards the cell in the thermal column. After flowing through the cell, the solution is routed through a section of tubing that is coiled and submerged in a room temperature water bath to bring down the solution's temperature. The water then passes through a capillary tube, dropping the pressure from >25 MPa to <1.5 MPa, before it flows into the glass sparging apparatus. A back pressure regulator relief valve (model Idex P-880) is included in the line between the bath and the capillary tube to prevent overpressurization of the cell beyond 28 MPa whenever heating a fresh volume of room temperature water.



Figure 2. Schematic of the system used for flowing experimental solution, containment and heating during irradiation, collection, and sparging to quantify dissolved gas content.

As part of each daily start-up procedure, the cell flow system is flushed with at least 500 mL of freshly prepared solution flowed through both paths to replace any impure solution remaining from previous runs. Once the reactor has achieved the desired power output level, a fresh volume of solution is flowed into the cell. The total volume of the flow system from the exit port of the syringe pump, through the cell, to the glass sparging apparatus inlet is estimated to be approximately 100 mL; therefore, for each instance when loading a new volume to be irradiated and/or when extracting an irradiated volume for testing, a total volume of 120 mL is made to flow to ensure full quantitative transfer.

The experimental procedure is based on a method developed originally by Janik et al. in 2007, as well as a more-specialized method for this reaction used by Dietz et al. in 2021 (Janik,

et al., 2007; Dietz, et al., 2021). After a sample volume has been irradiated and is ready for analysis, a new sample volume is pumped into the cell to simultaneously extract the entire irradiated volume into the glass sparging apparatus. The sparger is routed to allow a continuous stream of argon gas (99.999 % purity, Airgas Inc.), monitored by a flow controller (model Masterflex 32907-59), flowing either through a bypass route or bubbling up through the column of irradiated solution to sparge out the dissolved hydrogen and nitrogen. This mixed gas exits out the sparger, flows through two parallel Restek Molecular Sieve S-Traps to remove moisture, and passes an Inficon Transpector 2 Compact Process Monitor Residual Gas Analyzer (RGA) mass spectrometer for analysis. The RGA measures the ion current of select masses (amu 2, 28, and 32 for hydrogen, nitrogen, and oxygen, respectively) as a function of time. Integration of each gas's ion current signal peak area is proportional to the concentration of that gas dissolved within the solution.

Measuring the proportionality constant as a value of peak-area-per-mole of select gas is carried out daily. A volume of DI water is saturated with a gas mixture of (5.0 ± 0.1) % hydrogen and (5.0 ± 0.1) % nitrogen in argon (Airgas Inc). Saturated solution is drawn into the syringe pump, and select volumes (reported by the ISCO syringe controller to 0.1 cm³ accuracy) are flowed into the glass sparging apparatus. Volumes from 40 mL to 120 mL of solution are collected and sparged, and the resulting H₂ and N₂ signals are integrated to establish a linear relationship between the area of the signal peaks and the concentration of both gases. The concentration of each gas is expected to vary day-to-day due to temperature fluctuations in the experimental facility and so is calculated by Henry's Law. The values for the Henry's Law constant and their temperature dependence were obtained from the NIST Chemistry WebBook,

10

SRD 69, for both hydrogen gas and nitrogen gas (Lemmon, et al., 2021). A set of typical calibration signals is shown in Figure 3.



Figure 3. RGA ion current signal peaks for hydrogen (red) and nitrogen (green) for typical calibration sample volumes of 40mL, 80mL, and 120mL, with the 120mL sample containing 4.83 μ mol of H₂ and 4.09 μ mol of N₂. Linearity is demonstrated in the upper left corners. Error bars of the signal integration are smaller than the size of the points.

2.5 Neutron Flux Density

For irradiated samples, after analysis of all dissolved gases, the entire sample volume is collected and diluted to precisely 500 mL in a Marinelli beaker for ²⁴Na activation analysis to determine the total neutron exposure sustained by the sample. Activity in the Marinelli beaker is counted for 30 minutes on a Canberra high purity germanium (HPGe) detector model operating at 30 percent efficiency, using a Lynx Multichannel Analyzer, and analyzed using GenieTM 2000/Apex® spectroscopy software. ²⁴Na activity is based on a weighted average of the area under the peak located at 1368 keV. Based on the total ²⁴Na produced during the irradiation (roughly 1 μCi is generated per hour of exposure for the concentrations of sodium ion used), we calculate the average neutron flux density within the solution volume:

$$\Phi_{n} = \frac{A_{c} \ e^{\lambda_{Na-24} \,\Delta t}}{\sigma_{Na-23} \ (1 - e^{-(\lambda_{Na-24} \,t_{irr})}) c_{Na-23} V_{irr} N_{A}} \tag{1}$$

where Φ_n is the neutron flux density (s⁻¹ cm⁻²), A_c is the counted activity of ²⁴Na (Bq), λ_{Na-24} is the decay rate of ²⁴Na (1.287 × 10⁻⁵ s⁻¹), Δt is the decay time from the end of irradiation to the end of activation counting (s), σ_{Na-23} is the thermal absorption cross section of ²³Na ((5.33 × 10⁻²⁹ m² (0.533 barn)), t_{irr} is the irradiation duration (s), c_{Na-23} is the concentration of ²³Na (mol/L), V_{irr} is the volume of irradiated solution (L), and N_A is Avogadro's number. It is assumed the flux density is near-constant during the course of each sample's irradiation, and this is confirmed via the power trend logs from each day (an example is shown in Figure 1).

2.6 Gamma

The radiation spectrum in the RINSC thermal column initially was simulated using MCNP 6.2 (Werner, 2017) applied to a geometrically-simplified model of the experimental flowcell. (Monte Carlo N-Particle® or MCNP® is a general-purpose Monte Carlo radiation-transport code designed to track many particle types over broad ranges of energies, maintained by the Radiation Safety Informational Computational Center (RSICC) at Oak Ridge National Laboratory.) The MCNP simulation results showed for the zirconium and titanium alloys that the percentage of energy deposited in the boric acid solution from photons alone is 2.0% and 3.1%, respectively, of the total energy deposited by photons, alpha particles, and heavy ions.

The contribution to H_2 production both from the thermal column's low gamma radiation background and from prompt fluorescence gamma of the flowcell materials (titanium, Zircadyne 702 alloy, and stainless steel) has been measured to correct the ¹⁰B fission results. A zero-boron "blank" solution was used, containing similar concentrations of ²³Na for an *in-situ* neutron fluence dosimeter as in the boron experimental solutions, but with the addition of 1.0–4.0 mM isopropyl alcohol (70% in purified water, Pro Advantage, USP Grade) as a scavenger of the hydroxyl radical 'OH and 'H atom. Solution volumes were mixed, deaerated with the 2.5% N₂O in argon gas mixture, exposed for similar durations, and analyzed all in the same procedure as for the boron solutions. The chemistry induced by the gamma is very well known (Spinks & Woods, 1990; Elliot & Bartels, 2009; Sterniczuk, et al., 2016), producing e_{aq}^- , 'H atoms, and 'OH radicals which can react quantitatively with the scavengers as follows:

1)
$$N_2 O + e_{aq}^- + H_2 O \to N_2 + {}^{\circ}OH + OH^ G(e_{aq}^-) = 0.26 \,\mu \text{mol/J}$$

2)
$$(CH_3)_2CHOH + H \rightarrow H_2 + (CH_3)_2 COH \qquad G(H) = 0.062 \,\mu mol/J$$

3)
$$(CH_3)_2CHOH + {}^{\bullet}OH \rightarrow H_2O + (CH_3)_2 {}^{\bullet}COH$$
 $G(OH) = 0.26 \,\mu\text{mol/J}$

4)
$$(CH_3)_2 COH + (CH_3)_2 COH \rightarrow (CH_3)_2 CHOH + (CH_3)_2 CO$$

We therefore expect in this system a measured $g(H_2) = G(H) + G(H_2) = 0.062 + 0.047 = 0.109$ μ mol/J and $g(N_2) = G(e_{aq}) = 0.26 \mu$ mol/J solely from the low LET gamma.

In analyzing the background measurements, we were surprised to find in several repeat experiments that the ratio of N_2/H_2 signals is ca. 4–8 in these solutions rather than the expected ratio of $g(N_2)/g(H_2) = 2.4$. After some consideration, we realized the additional N_2 comes from a reaction of the isopropanol radicals with N_2O as:

5)
$$N_2O + (CH_3)_2 \cdot COH \rightarrow N_2 + \cdot OH + (CH_3)_2CO$$

Note that reaction (5) produces another ${}^{\circ}OH$ radical, meaning the N₂ is released in a chain reaction, with $(CH_3)_2 {}^{\circ}COH$ as the carrier. The isopropanol radicals are well-known to be powerful reducing species, but typically the rate constant for reduction of the N₂O is too low to be competitive with the self-disproportionation reaction (4) in a gamma source (several Gy per second dose rate). In the present case of several Gy per hour dose rate, the second-order selfdisproportionation is slow, and reaction (5) becomes important. We estimate a reaction rate on the order of $k_5 = 1 \times 10^3 M^{-1} s^{-1}$ can explain our observation. It means we should use the measured value for $g(H_2)$ for our gamma correction rather than the measured value of $g(N_2)$.

3 Results

Experimental runs were commonly performed at a target temperature on a given day, with conditions typically repeated for three measurements. A typical room temperature H₂ signal from the RGA is shown in Figure 4. The concentration of H₂ sparged out of each irradiated boric acid solution sample is calculated based on the daily calibrations, as described previously. The G-value for H₂ by the ¹⁰B(n, α)⁷Li reaction is directly calculated from the total quantity of H₂ present divided by the energy deposited in the solution by the reaction's product ions.

The high LET ions have a total of 2.793 MeV for 6% of the neutron capture events, and 94% of the events yield 2.314 MeV along with a 0.48 MeV gamma (Auden, et al., 2019). This gamma has a mean free path estimated to be 30 cm through water, which agrees with our simulation results showing very little absorbed dose from this gamma. Therefore it is omitted from the G-value calculation presented here, giving a weighted average of 2.343 MeV deposited in the solution per ${}^{10}B(n,\alpha)^{7}Li$ neutron capture fission event.

The thermal neutron capture cross section of boron-10 is $3.835 \times 10^{-25} m^2$ (3835 barn) (Sauerwein, et al., 2012), and the boric acid used in these experiments was confirmed via mass spectrometry to contain the natural isotopic fraction (0.199) (De Laeter, et al., 2003) of ¹⁰B, within measurement uncertainties. Therefore, our effective thermal neutron capture cross section is $7.63 \times 10^{-26} m^2$. The number of ¹⁰B neutron capture fission events can be calculated using the average neutron flux density obtained from the sodium-23 activation analysis by the following equation:

$$N_{Evt} = t_{irr} \Phi_n \sigma_{B-nat} c_{B-nat} V_{irr} N_A$$
(2)

where N_{Evt} is the number of ¹⁰B neutron capture events, t_{irr} is the duration of irradiation, σ_{B-nat} is the thermal absorption cross section of natural boron, and c_{B-nat} is the concentration of natural boron. This value can then easily be used to calculate the energy deposited in the solution by the ¹⁰B(n, α)⁷Li event. Using equations (1) and (2), the equation for G(H₂) becomes:

$$G(H_2) = \frac{\sigma_{Na-23} c_{Na-23}}{\sigma_{B-nat} c_{B-nat}} \times \frac{\left(1 - e^{-(\lambda_{Na-24} t_{irr})}\right)}{t_{irr} A_c e^{\lambda_{Na-24} \Delta t}} \times \frac{(H_2)}{E_{Evt}}$$
(3)

where (H_2) is the amount of hydrogen detected by the mass spectrometer (moles), and E_{Evt} is the energy deposited in solution per neutron fission event (3.754 × 10⁻¹³ J).

3.1 Gamma

MCNP simulations support the assumption that all gamma dose to the water results from neutron-capture fluorescence in the surroundings (none directly from the core) and thus is proportional to the measured neutron flux density in the sample. Based on the total content of hydrogen gas within an irradiated "gamma blank" sample, the total dose rate of gamma radiation can then be calculated by the following equation:

$$D_{\gamma} = \frac{(H_2)}{V_{Irr} \times g_{\gamma}(H_2) \times \rho_{H_2O}(T) \times t_{Irr}}$$
(4)

where D_{γ} is the gamma dose rate (Gy/hr), (H_2) is the amount of dissolved hydrogen (mol), V_{Irr} is the volume of the irradiated solution (L), $g_{\gamma}(H_2)$ is the g-value of molecular hydrogen for gamma at room temperature (0.109 µmol/J), $\rho_{H_2O}(T)$ is the density of water at a given temperature (kg/L), and t_{Irr} is the duration of irradiation (hr). The density of water must be taken into account here (Lemmon, et al., 2021); and although there is a corresponding change with temperature in the concentrations of boron and sodium in the boric acid experiment, their ratio remains constant and so the calculated yield of H₂ per ¹⁰B(n, α)⁷Li event is independent of the density of water. Figure 4 shows a plot of the raw signal obtained by the RGA mass spectrometer from a gamma blank sample compared to a typical boron sample. The gamma blank was irradiated for 120 minutes, whereas the boron sample was irradiated for 60 minutes. The extended duration used for gamma blanks was necessary to obtain sufficient S/N for dependable analysis, which demonstrates how minor the H₂ contribution from gamma fluorescence is here.



Figure 4. Comparison of the hydrogen signals obtained in a sample for measuring gamma contribution (green) and an experimental boron sample (black), both irradiated at room temperature. The gamma blank signal has been given an offset of -0.05nA.

Over the course of this several-month experimental campaign, the configuration of the flowcell, its containment, and the surroundings were altered in minor aspects (such as the type of heating tape used, and other assorted materials) as well as significantly (such as alternating the tubing material between Zircadyne and titanium). Therefore, several measurements were made to evaluate the gamma background once the cell was configured in a particular arrangement and continual experimentation was performed. Figure 5 displays the ratio of gamma dose (measured by generated hydrogen content) to the neutron flux density the sample experienced (measured by sodium activation) for the three main configurations of the flowcell used. According to this

evaluation, at room temperature D_{γ} is calculated to be 4.3 Gy/hr to 5.2 Gy/hr for the Zircadyne 702 cell material, and ca. 7.9 Gy/hr with the Grade 2 Titanium cell material.



Figure 5. Gamma dose rate versus neutron flux density, calculated from quantity of hydrogen produced and level of sodium activity, repectively, for different flowcell configurations throughout the experimental campaign: early experiments using Zircadyne cell (blue), midway experiments using titanium cell (black), later experiments again using Zircadyne cell (red). Error bars for gamma doserate are smaller than the size of the points.

In the boric acid experiments, the 'H reacts with H_2O_2 product or with NO_2^- and does not contribute to the measured H_2 level. There was no change with or without the NO_2^- scavenger.

Given the gamma dose rate deduced from the room temperature alcohol "blank" experiments, we use the following published equation (Elliot & Bartels, 2009) to calculate the radiation yield of H_2 produced by gamma background as a function of temperature:

$$G(H_2) = 0.419 + 8.712 \times 10^{-4} T - 4.971 \times 10^{-6} T^2 + 1.503 \times 10^{-8} T^3$$
 (5)

where *T* is the temperature of the aqueous matrix (°C) and $G(H_2)$ is in units of molecules/100eV (we multiply by 0.1036 for conversion to units of µmol/J). This G-value calculation is used in conjunction with equation (4) to compute the value of H₂ for subtraction from the raw measured quantity, where over the temperature range this gamma contribution accounted for 2–6% of the total hydrogen detected.

3.2 Corrosion

Both alloys used for flow cell tubing in this experiment are known to undergo some degree of corrosion in water with the stoichiometry indicated:

6) $Zr + 2H_2O \rightarrow ZrO_2 + 2H_2$ 7) $Ti + 2H_2O \rightarrow TiO_2 + 2H_2$

This oxidative corrosion process typically leads to the formation of a passivation layer of metal oxide (Was & Allen, 2019). The passive layer still has a non-zero corrosion rate, but the production of hydrogen becomes small.

The first corrosion cell used, made from Zircadyne 702, was found to corrode severely above 175 °C. The large background corrosion H₂ signal made it impossible to use. A backup

titanium tubing cell was substituted, and after conditioning for several days, reactor experiments could be carried out. On a given day, prior to reactor power-up, a sample of the boric acid solution was introduced into the titanium cell while it is held at a target temperature and 250 bar. This "corrosion blank" is kept in the cell for ca. 1 hour, then analyzed to quantify the hydrogen produced solely via corrosion during that time. A second blank is taken to confirm that the corrosion rate is constant. This amount of H₂ at the target temperature is then subtracted as "corrosion yield" from the raw hydrogen content measured in samples irradiated by neutrons at the same cell temperature later in the day.



Figure 6. Example hydrogen signals for a corrosion blank (red) and experimental sample (black), both obtained at 300 °C over a one-hour duration. The corrosion signal has been given an offset of -0.05nA.

Figure 6 compares the signals of a corrosion blank and a boron experiment sample, where both were heated to 300 °C and held at 250 bar for one hour. Experiments at 300 °C used half of the typical concentration of boric acid (0.15 M) to more accurately resemble the concentrations used in PWRs while running at this temperature. With the titanium cell, the amount of hydrogen produced in "corrosion blank" samples compared to the total hydrogen detected in irradiated samples was ca. 17 % at 300 °C, 8 % at 250 °C, and only 1 % at 200 °C.

3.3 Final Results of G(H₂)

A linear plot of the hydrogen content detected versus calculated energy deposited in solution samples at room temperature is shown in Figure 7, where the slope fit by linear regression to the data is equal to a G-value for H₂ of $(0.162 \pm 0.007) \mu mol/J$. This value has been corrected for the small amount of H₂ produced by the evaluated gamma background. No significant effect was found at room temperature by the inclusion of ⁷LiOH to balance the pH of the boric acid solution to 7. Regardless, the ⁷LiOH was included in all sample solutions for all experiments at temperatures above 200 °C to more closely match PWR chemistry conditions.



Figure 7. Plot of the room temperature measurements for calculating the G-value of hydrogen generation by the ${}^{10}B(n,\alpha)^{7}Li$ reaction after corrections made for gamma background.

The error bars displayed in Figure 7 are associated with the experimentally estimated uncertainty in the measurements of sodium activation and of hydrogen content. The reports generated by the Genie gamma spectroscopy software have uncertainty values (σ_{Act}) for the detected levels of activated sodium-24, and these are reflected in the horizontal error bars for the values of energy deposited in the solution as calculated using Equation (2) and the known product energy of each ${}^{10}B(n,\alpha){}^{7}Li$ event. A standard deviation (σ_{H2}) is computed for the proportionality constant of the three hydrogen calibration measurements performed each day, and this is reflected in the vertical error bars for the values of hydrogen gas detected by the RGA

mass spectrometer on that day. The sodium activation uncertainties reported throughout the entirety of this experiment average 3.7 % of their measured value, and all σ_{H2} average 1.6 % of their measured proportionality constant.



Figure 8. Plot of all calculated G-values for all measurements on the ${}^{10}B(n,\alpha)^{7}Li$ reaction taken in this work.

The G-values have been calculated from the hydrogen analysis and sodium activation reports as explained in the discussion for Equation (3), and all results have been plotted in Figure 8. The standard deviation error bars in Figure 8 for the G-values were calculated as

$$\sigma_{G(H_2)} = G(H_2) \times \sqrt{(\sigma_{H_2}/H_2)^2 + (\sigma_{Act}/Act)^2}$$
(6)

However, as the data in these two figures indicates, these calculated errors do not account for the spread of measured values and thus are considered underestimations of the true total experimental uncertainty. Therefore, each G-value is treated equivalently and an average G-value at each temperature is computed. The error bars are indicative of one standard deviation for the G-values at that temperature. The error for the temperature was estimated as ± 5 °C based on the accuracy of our thermocouples determined by observing their behavior over the course of this experimental campaign. The final G-values for H₂ generated by the recoil ions of the ¹⁰B(n, α)⁷Li reaction in aqueous solution at 25 MPa, after correction for the excess H₂ contributed by gamma background and corrosion, are given in Table 1. Uncertainties given are k = 1, i.e., they define intervals around the central value within which the actual value is believed to lie with a level of confidence of approximately 68 %.

Table 1. G-values for H_2 from the ${}^{10}B(n,\alpha)^7Li$ reaction in aqueous solution at 25 MPa for a temperature range up to 300 °C.

Τ	G(H ₂) (µmol/J)	G(H ₂) (#/100eV)	H ₂ molecules/Event
20	0.162 ± 0.007	1.56 ± 0.06	$36,600 \pm 1,500$
60	0.169 ± 0.007	1.63 ± 0.07	$38,200 \pm 1,500$
80	0.165 ± 0.008	$\boldsymbol{1.59 \pm 0.08}$	$37,200 \pm 1,900$
120	$\boldsymbol{0.170 \pm 0.009}$	$\boldsymbol{1.64 \pm 0.09}$	$38,500 \pm 2,100$
160	0.170 ± 0.009	1.64 ± 0.09	$38,400 \pm 2,000$
190	$\boldsymbol{0.180 \pm 0.006}$	$\boldsymbol{1.73\pm0.06}$	$40,\!600 \pm 1,\!400$
250	0.184 ± 0.005	$\boldsymbol{1.78 \pm 0.05}$	$41,\!600 \pm 1,\!100$
300	0.162 ± 0.005	$\boldsymbol{1.57\pm0.05}$	$36,700 \pm 1,100$

4 Discussion

As previously mentioned, there is a scarcity of radiation yield measurements for the ${}^{10}B(n,\alpha)^7Li$ reaction in aqueous solutions, particularly the G(H₂) and G(H₂O₂) needed for reactor modeling. Barr and Schuler reported yields for G(•H), G(•OH), and G(H₂O₂) by the ${}^{10}B(n,\alpha)^7Li$ reaction in an aqueous 0.8N sulfuric acid solution using spectrophotometric detection of the oxidation of ferrous ions (Fe²⁺) (Fricke dosimetry) and reduction of ceric ions (Ce⁴⁺) both with and without the presence of dissolved oxygen (Barr and Schuler, 1956; Schuler and Barr 1959). Later, LaVerne and Schuler reported on the same product yields in the Fricke dosimeter solution generated from individual accelerator-produced alpha particles and ${}^7Li^{1+}$ ions, and their results are in good agreement with the yields by Barr and Schuler (LaVerne & Schuler, 1987).

In the precursor to the present work, Dietz et al. reported on the molecular hydrogen yield by the ${}^{10}B(n,\alpha)^7Li$ reaction in a neutral-pH boric acid solution at room temperature (Dietz, et al., 2021). This experiment can be faulted for failure to anticipate the significant gamma background from neutron capture by titanium in the cell block used. In order to recover a G(H₂) number, MCNP simulations were carried out, which indicated that 29 % of all energy deposited was from ⁴⁸Ti neutron activation gamma fluorescence. To add uncertainty, there was no adequate scavenger for 'H atoms or 'OH radicals in this experiment. Our present G(H₂) result of (0.162 ± 0.007) µmol/J replaces the original report of (0.12 ± 0.01) µmol/J.

In 2006, Christensen produced a report on radiolysis in nuclear reactors, including a compilation and assessment on the available values of important parameters for computer simulations of these processes. For H₂ production by ${}^{10}B(n,\alpha)^{7}Li$, he lists experimental and simulation results at room temperature from several sources (Jenks & Griess, 1967; Lefort, 1958)

and provides his own recommended G-value of 0.145 μmol/J (Christensen, 2006), as well as two calculated estimates for ca. 300 °C (Bjergbakke, et al., 1984; Lundgren, et al., 2004).

To our knowledge, only these simple estimates of $G(H_2)$ from ¹⁰ $B(n,\alpha)^7$ Li were available for high temperature PWR reactor simulations until Monte Carlo track chemistry calculations of the fission event in aqueous solution were carried out at Sherbrooke University up to 350 °C (Islam, et al., 2017). The Sherbrooke results are plotted in Figure 9 along with our averaged experimental values and all earlier values. The simulations using experimental reaction rate numbers compiled in a review by Elliot and Bartels (Elliot & Bartels, 2009) are plotted as green squares. Agreement is good up to 150 °C. The simulated yield decreases between 150 °C and 250 °C because the measured rate constant k₈ for bimolecular reaction of hydrated electrons,

8)
$$e_{aa}^- + e_{aa}^- + 2H_2O \rightarrow H_2 + 2OH^-$$

decreases catastrophically in this temperature range, as reported independently by three separate research groups (Elliot & Bartels, 2009). It was expected that this reaction should have significant probability in both high LET tracks (Swiatla-Wojcik & Buxton, 1998; Islam, et al., 2017) and low LET spur recombination (Swiatla-Wojcik & Buxton, 1995; Sanguanmith, et al., 2011) and the same prediction of dip in the H₂ yield above 150 °C is made in both low- and high-LET situations. With our new high temperature measurements of high LET ¹⁰B(n, α)⁷Li radiolysis we see (c.f. Figure 9) it is not found experimentally in either case. (Elliot & Bartels, 2009; Sterniczuk, et al., 2016). Jay-Gerin and coworkers have repeatedly suggested (Sanguanmith, et al., 2011; Butarbutar, et al., 2013; Islam, et al., 2017) that the k₈ reaction rate decrease only occurs in alkaline solutions where the kinetics measurements are made, but not in neutral pH conditions. The second set of simulations plotted as blue circles in Figure 9 were

calculated (Islam, et al., 2017) by extrapolating the rate constant k_8 from low temperature measurements to high temperature using the Arrhenius law as originally suggested by Elliot (Elliot, 1994). This undeniably produces better agreement between the Sherbrooke simulation and experimental G(H₂) values.



Figure 9. Plot of $G(H_2)$ values from ${}^{10}B(n,\alpha)^7Li$ reaction reported in this work, along with comparable results from other works.

Unfortunately the suggestion of Jay-Gerin and coworkers (Sanguanmith, et al., 2011; Butarbutar, et al., 2013; Islam, et al., 2017) is both non-physical from the standpoint of reaction rate theory, and not supported by experiment. Imagine a high temperature (e.g., 200 °C) encounter of two solvated electrons. According to Jay-Gerin's suggestion, reaction will occur in the neutral solution, where virtually no ions are present, but not in the alkaline solution, where a mere 1×10^{-4} M concentration of KOH (as used in kinetics experiments to scavenge protons (Marin, et al., 2007)) can somehow completely prevent reaction. What can possibly be different at such low concentration? Ionic strength effects are small. Three body effects are impossibly rare except for the solvent. One might postulate ion pairing of the electrons with K⁺ or Na⁺ ions to dramatically decrease the diffusion and reaction rates. But then other diffusion-limited reactions of e_{aq}^- would show similar effects, and addition of inert salts would produce the same result. In fact, the G(H₂) has been measured at high temperature under alkaline 1x 10⁻³ mol/L NaOH conditions for low LET electron beam radiation (Sterniczuk, et al., 2016). There is no difference from neutral pH yield measurements. The "dip" above 150 °C is missing even in alkaline solution where k₈ certainly drops.

If we accept that reaction rate k_8 really does decrease above 150 °C, the lack of any "dip" in the H₂ yield of Figure 9 means that reaction (8) is not a very important source of H₂ in high LET radiolysis at 150 °C, and the Sherbrooke model (Islam, et al., 2017) is incorrect. Using high scavenger concentrations for pre-solvated electrons, it has been possible to separate the prompt physico-chemical G₀(H₂) from overall escape yields G_{esc}(H₂) which also includes diffusive track recombination (Sterniczuk & Bartels, 2016). Already at room temperature, it is demonstrated that for high LET alpha particles G₀(H₂) is 0.1 µmol/J and overall G_{esc}(H₂) is ca. 0.16 µmol/J (LaVerne & Pimblott, 2000; Sterniczuk, et al., 2016). For low LET *e*-beam radiation, it was found that G₀(H₂) is responsible for significantly increasing the overall G_{esc}(H₂) with temperature (Sterniczuk, et al., 2016). One could expect that at higher temperature, the already-large fraction of G₀(H₂) for alpha radiation would become even higher, and the fraction of G_{esc}(H₂) coming

28

from reaction (8) drops significantly below 10 %. In this case any "dip" could be below our present experimental detection limit.

5 Conclusion

The present work presents the first direct measurements for $G(H_2)$ from the ${}^{10}B(n,\alpha)^7Li$ fission process as a function of temperature up to Pressurized Water Reactor conditions. The temperature dependence is quite weak. Simple estimates used previously for this quantity in reactor models were not too far off. Detailed Monte Carlo simulations previously carried out at Sherbrooke University (Islam, et al., 2017) predicted a dip in $G(H_2)$ above 150 °C due to the catastrophic decrease in reaction rate k₈ for bimolecular recombination of e_{aq}^- . Based on all of the experimental evidence we are forced to the conclusion that the importance of this reaction in spur and track recombination is significantly overestimated in the Sherbrooke model.

Disclaimer

Certain commercial equipment, instruments, or materials are identified in this paper in order to specify the experimental procedure adequately. Such identification is not intended to imply recommendation or endorsement by NIST, nor is it intended to imply that the materials or equipment identified are necessarily the best available for the purpose.

Author contributions

The manuscript was written through contributions of all authors. All authors have given approval to the final version of the manuscript.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

Cameron Goodwin and Sangho Nam were supported by the Rhode Island Nuclear Science Center. Alan Thompson was supported by the Physical Measurements Laboratory of NIST. David Bartels and Steven Guerin were supported by the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences under award number DE-FC02-04ER15533. This is manuscript number 5393 of the Notre Dame Radiation Laboratory.

References

- Auden, E. et al., 2019. Thermal neutron-induced single-event upsets in microcontrollers containing boron-10. *IEEE Transactions on Nuclear Science*, 67(1), pp. 29-37.
- Barr, N. F. & Schuler, R. H., 1959. The dependence of radical and molecular yields on linear energy transfer in the radiation decomposition of 0.8 N sulfuric acid solutions. *Journal* of Physical Chemistry, 63(6), pp. 808-812.
- Bartels, D., Henshaw, J. & Sims, H., 2013. Modelling the Critical Hydrogen Concentration in the AECL Test Reactor. *Radiation Physics and Chemistry*, Volume 82, pp. 16-24.
- Bjergbakke, E., Sehested, K., Rasmussen, O. & Christensen, H., 1984. Input Files for Computer Simulation of Water Radiolysis, DK 4000 Roskilde, Denmark: Risø National Laboratory.
- Brown, D. A. et al., 2018. ENDF/B-VIII.0: The 8th Major Release of the Nuclear Reaction Data Library with CIELO-project Cross Sections, New Standards and Thermal Scattering Data. *Nuclear Data Sheets*, 2, Volume 148, pp. 1-142.
- Butarbutar, S. L. et al., 2013. On the Temperature Dependence of the Rate Constant of the Bimolecular Reaction of Two Hydrated Electrons. *Atom Indonesia*, Volume 39, pp. 51-56.
- Butson, M. & Niroomand-Rad, A., 2017. Historical Background, Development, and Construction of Radiochromic Films. In: *Radiochromic Film.* s.l.:CRC Press, pp. 7-32.
- Carlson, A. D., 2011. The Neutron Cross Section Standards, Evaluations and Applications. *Metrologia*, 12, 48(6), p. S328.
- Carlson, A. D. et al., 2018. Evaluation of the Neutron Data Standards. *Nuclear Data Sheets*, 2, Volume 148, pp. 143-188.
- Christensen, H., 1995. Remodeling of the oxidant species during radiolysis of hightemperature water in a pressurized water reactor. *Nuclear Technology*, 109(3), pp. 373-382.

- Christensen, H., 2006. *Fundamental Aspects of Water Coolant Radiolysis,* Stockholm, Sweden: Swedish Nuclear Power Inspectorate.
- Crow, M. et al., 1995. Thermal neutron measurements of the Rhode Island Nuclear Science Center reactor after conversion to a compact low enriched uranium core. *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment*, 365(2-3), pp. 433-445.
- De Laeter, J. et al., 2003. Atomic weights of the elements. Review 2000 (IUPAC Technical Report). *Pure and Applied Chemistry*, 75(6), pp. 683-800.
- Dietz, T. C. et al., 2021. H₂ production in the ¹⁰B(n,α)⁷Li reaction in water. *Radiation Physics and Chemistry*, 3, Volume 180, p. 109319.
- Elliot, A., 1994. Rate constants and G-values for the simulation of the radiolysis of light water over the range 0-300 deg C (No. AECL--11073). *Atomic Energy of Canada Ltd..*
- Elliot, A. & Bartels, D., 2009. *The reaction set, rate constants and g-values for the simulation of the radiolysis of light water over the range 20 deg to 350 deg C based on information available in 2008.* s.l.:Atomic Energy of Canada Limited.
- Islam, M. M. et al., 2017. Monte Carlo track chemistry simulations of the radiolysis of water induced by the recoil ions of the 10B(n,α)7Li nuclear reaction. 1. Calculation of the yields of primary species up to 350 °C. *RSC Advances*, 7(18), pp. 10782-10790.
- Janik, D., Janik, I. & Bartels, D. M., 2007. Neutron and β/γ radiolysis of water up to supercritical conditions. 1. β/γ yields for H2, H atom, and hydrated electron. *Journal* of Physical Chemistry A, 8, 111(32), pp. 7777-7786.
- Jenks, G. H. & Griess, J. C., 1967. *Water Chemistry in Pressurized and Boiling Water Power Reactors (No. ORNL-4173),* Oak Ridge, Tennessee: Oak Ridge National Laboratory.
- Kusumoto, T. & Ogawara, R., 2019. Radiation Chemical Yield of Hydroxyl Radicals for Accelerator-based Boron Neutron Capture Therapy: Dose Assessment of 10B(n,α)7Li Reaction Using Coumarin-3-Carboxilic Solution. *Radiation Research*, 5, 191(5), pp. 460-465.

- Kusumoto, T. et al., 2020. Water radiolysis with thermal neutrons, fast neutrons and contamination gamma rays in the accelerator based thermal neutron field: Time dependence of hydroxyl radical yields. *Radiation Physics and Chemistry,* Volume 174, p. 108978.
- Laverne, J. A., 2000. Track Effects of Heavy Ions in Liquid Water. *Radiation Research*, 153(5), pp. 487-496.
- LaVerne, J. A., 2004. Radiation Chemical Effects of Heavy Ions. In: *Charged Particle ad Photon Interactions with Matter.* New York: Marcel Dekker, Inc., pp. 403-429.
- LaVerne, J. A. & Schuler, R. H., 1987. Radiation chemical studies with heavy ions: Oxidation of ferrous ion in the Fricke dosimeter. *Journal of Physical Chemistry*, 91(22), pp. 5770-5776.
- LaVerne, J. & Pimblott, S., 2000. New Mechanism for H2 Formation in Water. *Journal of Physical Chemistry A*, 104(44), pp. 9820-9822.
- Lefort, M., 1958. Radiation Chemistry. *Annual Review of Physical Chemistry*, 9(1), pp. 123-156.
- Lemmon, E., Bell, I., Huber, M. & McLinden, M., 2021. Thermophysical Properties of Fluid Systems. In: W. Mallard & P. Linstrom, eds. *NIST Chemistry WebBook, NIST Standard Reference Database Number 69.* Gaithersburg, MD 20899: National Institute of Standards and Technology.
- Lin, C., 2000. Hydrogen water chemistry technology in boiling water reactors. *Nuclear Technology*, 130(1), pp. 59-70.
- Lundgren, K., Wijkström, H. & Wikmark, G., 2004. *Recent developments in the LwrChem radiolysis code.* Intl. Conf. water Chem. Nucl. Reactor Systems, San Fransisco.
- Macdonald, D., 1992. Viability of hydrogen water chemistry for protecting in-vessel components of boiling water reactors. *Corrosion*, 48(3), pp. 194-205.
- Marin, T. W. et al., 2007. Recombination of the Hydrated Electron at High Temperature and Pressure in Hydrogenated Alkaline Water. *Journal of Physical Chemistry A,* Volume

111, pp. 11540-11551.

Mitsui, j. & Siguyama, K., 1973. Neutron Thermalization in Graphite. *Journal of Nuclear Science and Technology*, 10(1), pp. 1-9.

Mozumder, A., 1999. Fundamentals of Radiation Chemistry. San Diego: Academic Press.

- NIST, 2022. *Atomic Weights and Isotopic Compositions for All Elements.* [Online] Available at: <u>https://physics.nist.gov/cgi-bin/Compositions/stand_alone.pl</u>
- Raiman, S., Bartels, D. & Was, G., 2017. Radiolysis driven changes to oxide stability during irradiation-corrosion of 316L stainless steel in high teperature water. *Journal of Nuclear Materials,* Volume 493, pp. 40-52.
- RIAEC, January 2017. Safety Evaluation Report: Renewal of the Facility Operating License for the Rhode island Nuclear Science Center Reactor. License No. R-95. Docket No. 50-193, Rhode Island Atomic Energy Commission: United States Nuclear Regulatory Commission, Office of Nuclear Reactor Regulation.
- Sanguanmith, S. et al., 2011. Low-linear Energy Transfer Radiolysis of Liquid Water at Elevated Temperatures up to 350 Degrees C: Monte-Carlo Simulations. *Chemical Physics Letters*, Volume 508, pp. 224-230.
- Sauerwein, W., Wittig, A., Moss, R. & Nakagawa, Y., 2012. Neutron Capture Therapy -Principles and Applications. Berlin Heidelberg (Germany): Springer-Verlag Berlin Heidelberg.
- Sekine, T. & Baba, H., 1980. A study of Reactor-Neutron-Induced Reactions: Double Neutron Capture Process and the Systematics of the (n,2n) Reaction, Japan: Japan Atomic Energy Research Institute.
- Spinks, J. & Woods, R., 1990. *An Introduction to Radiation Chemistry.* Third ed. s.l.:John Wiley & Sons, Inc..
- Sterniczuk, M. & Bartels, D. M., 2016. Source of Molecular Hydrogen in High-Temperature Water Radiolysis. *Journal of Physical Chemistry A,* Volume 120, pp. 200-209.

Sterniczuk, M. et al., 2016. Low LET radiolysis escape yields for reducing radicals and H2 in

pressurized high temperature water. *Radiation Physics and Chemistry*, Volume 121, pp. 35-42.

- Swiatla-Wojcik, D. & Buxton, G., 1995. Modeling of Radiation Spur Processes in Water at Temperatures up to 300-Degrees-C. *Journal of Physical Chemistry*, Volume 99, pp. 11464-11471.
- Swiatla-Wojcik, D. & Buxton, G., 1998. Modelling of linear energy transfer effects on track core processes in the radiolysis of water up to 300 degrees C. *Journal of the Chemical Society-Faraday Transactions*, Volume 94.
- Tehan, T., 2000. The Rhode Island Nuclear Science Center Conversion from HEU to LEU Fuel, Narragansett, RI: Rhode Island Nuclear Science Center.
- Was, G. & Allen, T., 2019. Corrosion Issues in Current and Next-Generation Nuclear Reactors. *Structural Alloys for Nuclear Energy Applications*, pp. 211-246.
- Werner, C., 2017. MCNP Users Manual-Code Version 6.2 (Report LA-UR-17-29981), New Mexico: Los Alamos National Laboratory.
- Yokohata, A. & Tsuda, S., 1974. A Solvated Electron Formed from the Water by the Irradiation of the Recoil Particles of 10B(n,a)7Li and 6Li(n,a)T. *Bulletin of the Chemical Society of Japan*, 47(11), pp. 2869-2870.