Design and control of the oxygen partial pressure of UO_2 in TGA using the humidification system

S. Lee, T. W. Knight*, and E. Roberts

*Nuclear Engineering, University of South Carolina, 300 Main Street, Columbia, SC 29208,
United States*

Abstract

The oxygen to uranium (O/U) ratio of $\text{UO}_{2\pm x}$ is determined by the oxygen content of the sample and is affected by oxygen partial pressure ($p\text{O}_2$) of the surrounding gas. Oxygen partial pressure is controllable by several methods. A common method to produce different oxygen partial pressures is the use of equilibria of different reaction gases. There are two common methods: $\text{H}_2\text{O}/\text{H}_2$ reaction and CO_2/CO reaction. In this work, $\text{H}_2\text{O}/\text{H}_2$ reaction using a humidifier was employed and investigated to ensure that this humidification system for oxygen partial pressure is stable and accurate for use in Thermogravimetric Analyzer (TGA) experiments with UO_2 . This approach has the further advantage of flexibility to make a wide range of oxygen partial pressure with fixed hydrogen gas flow rate only by varying temperature of water in the humidifier. The whole system for experiments was constructed and includes the humidification system, TGA, oxygen analyzer, and gas flow controller. Uranium dioxide (UO_2) samples were used for experiments and oxygen partial pressure was measured at the equilibrium state of stoichiometric $\text{UO}_{2.0}$. Oxygen partial pressures produced by humidification (wet gas) system were compared to the approach using mixed dry gases (without humidification system) to demonstrate that the humidification system provides for more stable and accurate oxygen partial pressure control. This work provides the design, method, and analysis of a humidification system for oxygen partial pressure control in experiments with oxide nuclear fuels.

Keywords: *Humidification system; Oxygen partial pressure ($p\text{O}_2$); UO_2 ; $\text{H}_2\text{O}/\text{H}_2$ reaction; TGA; Oxygen analyzer*

*Corresponding author. Tel.: +1 803 777 1465.

Email: knighttw@cec.sc.edu

1. Introduction

A humidification system was designed to control oxygen partial pressure in experiments with uranium oxide nuclear fuels. The properties and thermochemistry of a uranium dioxide (UO_2) fuel is related to the oxygen pressure within the fuel element. The oxygen partial pressure or the oxygen potential in the UO_2 fuel is determined by fuel temperature and fission products in the UO_2 fuel. Advances in understanding of the thermochemistry of uranium oxide fuels is required for development of higher burnup fuels with greater fission product concentrations. Several methods have been used to measure oxygen potentials of the oxide fuels such as solid state EMF, Knudsen effusion, direct oxygen pressure measurement, and gas equilibrium with Thermogravimetric Analyzer (TGA) [1]. In this study, gas equilibrium in a TGA was augmented by the addition of a humidifier to the TGA system. The experimental setup consists of a humidifier, TGA, oxygen analyzer, and gas flow controller. The purpose of this assembled unit is for the simultaneous measurement of oxygen to uranium (O/U) ratio, oxygen partial pressure (p_{O_2}), and temperature. All sample mass change data are recorded by the TGA and all data of the oxygen content of the reaction gases are recorded by the oxygen analyzer. There are two common gas equilibrium methods to control the oxygen partial pressure. One method is the use of the $\text{H}_2\text{O}/\text{H}_2$ equilibrium and the other is the CO_2/CO equilibrium. Dry gas methods of $\text{H}_2\text{O}/\text{H}_2$ or CO_2/CO may be preferred for their simplicity requiring only the mixing of various gas compositions. Nevertheless, this work took advantage of the $\text{H}_2\text{O}/\text{H}_2$ reaction with the addition of a humidification system to vary the H_2O content by varying the temperature of the humidification system and thereby affecting the oxygen partial pressure. The reaction time of $\text{H}_2\text{O}/\text{H}_2$ to produce a given oxygen partial pressure is faster than that of CO_2/CO equilibrium and it is possible to produce a wide range of oxygen partial pressures with a fixed hydrogen gas flow rate and varying only temperature of the humidification system [2]. Other issues with the CO_2/CO method include fidelity requirements for flow control of the gases to yield a wide range of oxygen partial pressures and the general hazard concern over CO gas use. This modification to a TGA system is expected to provide for better control of oxygen partial pressure in nuclear fuel thermochemical studies.

2. Experimental Procedure

2.1. Experimental setup

Oxygen partial pressure yielded by the humidification system reacts with the uranium oxide sample and is measured by the zirconia oxygen sensor in the oxygen analyzer. Powder samples of uranium oxide were used for experiments and stoichiometry of the uranium oxide sample was unknown at the outset of each experiment. Samples were first oxidized to U_3O_8 and then reduced to UO_2 at several different temperatures. Oxygen partial pressure at each equilibrium state was monitored and all data such as temperature, mass change, oxygen partial pressure, and flow rate were collected. **Table 1** shows the summary of several experimental conditions applied in this work.

<Table 1>

Figure 1 shows the whole assembled experimental unit and Figure 2 shows details of the humidifier and TGA. As shown in Figure 2, hydrogen gas flowing through the humidity (water) bottle picks up water vapor that is carried downstream into the TGA. The equilibrium established between the hydrogen and water vapor at the temperature of the TGA establishes the oxygen partial pressure for the experiment.

<Figure 1>

<Figure 2>

When the humidification system is used, condensation of the water vapor has to be considered because condensed water could damage the TGA or Oxygen Analyzer. The humidity bottle was supplied by Fuel Cell Technology Inc. and dew point has been measured for varying flow rates and water bottle temperatures and is approximately 1 degree but less than 2 degrees below the water bottle temperature for highest flow rates of 250ml/min [3]. Heating tape is used to maintain the temperature in the gas line higher than the dew point temperature to prevent condensation of water.

2.2. Calculation of oxygen potentials and range for system operation

In this work, H₂O/H₂ gas mixture is used to create a range of oxygen partial pressures using hydrogen gas and water vapor added by a humidity bottle as an alternate method to gas mixing. The chemical reaction for the H₂O/H₂ gas mixture is as follow:

Oxygen partial pressure in this reaction is dependent on the equilibrium between hydrogen and water vapor, and can be calculated if partial pressure of H₂ and H₂O and temperature of humidity bottle are given. Water vapor in the humidity bottle has a specific water vapor pressure depending on water temperature. The water vapor pressure can be approximated from the saturated water vapor pressure based on manufacturer data showing the measured dew point is close to the water bottle temperature (i.e. 79.4°C for 80°C water bottle temperature at a typical 100 ml/min gas flow rate) [3]. The saturated water vapor pressure was taken from the water vapor pressure table shown in Table 2 [4].

<Table 2>

The standard Gibbs free energy change in the equilibrium state for the H₂O/H₂ gas reaction is expressed by the following Equation (1).

$$\Delta G^\circ = \Delta H^\circ - T\Delta S^\circ = -RT\ln(K) = -RT\ln\left(\frac{p_{H_2O}^2}{p_{H_2}^2 \times p_{O_2}}\right) \quad (1)$$

Then for a given temperature T in Kelvin, the known standard Gibbs free energy change, and H₂O and H₂ gas partial pressure, the achievable oxygen partial pressure with the humidification system was calculated. Table 3 shows the calculated oxygen partial pressure ranges in the furnace of TGA for different furnace temperatures, humidity bottle temperatures, and different gas mixtures. According to the Table 3, a range of oxygen partial pressures can be produced with fixed hydrogen gas flow rate by changing only the temperature of water in the humidifier. This demonstrates how flexible the humidification system is to control oxygen partial pressure.

<Table 3>

The results of Table 3 are sufficient to demonstrate the range of oxygen partial pressures possible. For a more accurate calculation of the oxygen partial pressure, it has to take impurities in the gas mixture into account and also the slight temperature dependence of ΔH and ΔS in Equation 1. The oxygen partial pressure at the TGA operating temperature was thus calculated by thermochemistry computer software (FactSage) before performing experiments in this work [5]. This is called the predicted oxygen partial pressure in this work. First, compositions of all mixed gases are given by the manufacturer. Then, each individual gas concentration is calculated depending on the Dalton's law which is equivalent to the gas partial pressure of each individual gas in the mixture. When this flowing dry gas passes through the water in the humidity bottle, it picks up water vapor as described earlier. So, the saturated water vapor pressure is added to the mixed dry gas pressure as an approximation to the concentration of H₂O in the flowing gas. Then, the calculated concentration of each component is equal to the mole fraction of each gas component. Second, these mole fractions of all components and the applied temperature of TGA

are input to the Equilibrium Module of FactSage which calculates the equilibrium oxygen partial pressure inside the TGA furnace. This predicted oxygen partial pressure is compared to the measured oxygen partial pressure from the oxygen analyzer in each experiment.

2.3. Measurement of pO_2

The oxygen analyzer is connected to the exhaust line from the TGA and measures oxygen concentration of the reaction gas from the TGA. This oxygen analyzer employs a zirconia (zirconium oxide) sensor. This oxygen sensor contains a small zirconia electrolyte tube, which is heated to about 650°C in order to conduct oxygen ions. This oxygen analyzer allows fast and accurate oxygen analysis over the full oxygen range from 1,000,000 ppm to 10^{-20} ppm. Sensor response time is typically 5 seconds at low oxygen concentration and 4 seconds at high oxygen concentration. The accuracy of the sensor is $\pm 1\%$ of the actual measured oxygen content and precision is better than $\pm 0.5\%$ [6].

After oxygen concentration is measured, the data is recorded. To calculate oxygen partial pressure in atmosphere (atm), the conversion of units is needed because the oxygen analyzer uses ppm for oxygen concentration and bar for the system pressure. For the conversion of units, Equation (2) is used.

$$pO_2(atm) = \left(\frac{O_2 \text{ ppm}}{1000000} \right) \times \left(\frac{1.0132 \text{ bar}}{\text{sensor internal pressure bar}} \right) \quad (2)$$

In Equation (2), the first term on the right side is the oxygen concentration in ppm and the second term in the right side is the ratio of the gas pressure outside versus inside of the oxygen sensor. Normally, internal pressure of the oxygen sensor is about from 0.001 to 0.01 bar higher than outside pressure of the oxygen sensor depending on the gas flow rate. Uncertainty for the measured oxygen partial pressure was calculated as $\pm 0.95\%$ of the measured oxygen concentration and oxygen sensor internal pressure by error propagation from Equation (2).

2.4. Measurement of O/U ratio

The flexibility of the humidification system to control oxygen partial pressure was demonstrated in a series of oxidation reduction experiments with uranium. The fundamental theory of the oxidation and reduction of a uranium oxide sample is based on the equilibrium state between the sample and oxygen partial pressure surrounding the sample. For a simple example, the reaction is as follow:

At this equilibrium state, oxygen potential of the uranium oxide sample is equivalent to the oxygen partial pressure of the reaction gas according to Equation (3) [7].

$$\Delta G_{O_2}^\circ = -RT \ln(K) = -RT \ln \left(\frac{a_{UO_2}}{a_U \times p_{O_2}} \right) = RT \ln(p_{O_2}) \quad (3)$$

where, UO_2 and U are unit activity of 1 and the activity of O_2 is equal to its partial pressure.

The O/U ratio in the single phase range of the stoichiometry is calculated at the completion of the TGA experiment. To establish a reference point for O/U ratio in oxidation/reduction experiments, each sample was first oxidized in air to U_3O_8 in equilibrium and noting its mass. Next the desired pO_2 is sought to reduce the sample to $UO_{2.0}$, the second equilibrium state, and the mass of $UO_{2.0}$ is noted. Several baseline tests without a sample were performed before an actual sample test was carried out for an accurate measurement. The result with the actual sample test in TGA shows a net mass change of the sample offset by a baseline test. There is no mass change of the sample at the equilibrium state, so that the horizontal TG line is observed on the TGA monitor. In order to decide the desired pO_2 to reduce the sample to $UO_{2.0}$, oxygen partial pressure range for $UO_{2.0}$ at a given temperature was used from Blackburn

(1973) [8]. A specific pO_2 in this range was chosen and the humidification system was set to reproduce this oxygen partial pressure according to the predicted value.

The O/U ratio calculation is expressed in Equation (4). Uncertainty of the calculated O/U ratio is related to the error of the oxygen fraction and that of the uranium fraction. This error descends from the uncertainty in the TGA mass balance. In this case, the uncertainty is calculated by error propagation of O/U expressed as Equation (5).

$$\frac{O}{U} = \frac{\frac{\text{actual weight of oxygen in a sample}}{\text{molecular weight of oxygen}}}{\frac{\text{actual weight of uranium in a sample}}{\text{molecular weight of uranium}}} \quad (4)$$

$$\Delta \frac{O}{U} = \left(\frac{O}{U} \text{ratio} \right) \times \sqrt{\left(\frac{\text{error of fraction}}{\text{oxygen fraction}} \right)^2 + \left(\frac{\text{error of fraction}}{\text{uranium fraction}} \right)^2} \quad (5)$$

Each sample has its own uncertainty ranging from ± 0.0012 to ± 0.0015 .

3. Results and Discussion

3.1. Oxygen potential and O/U ratio at the equilibrium state

Table 4 shows the results of four experiments at four different TGA temperature. The calculated sample O/U ratio and the measured oxygen partial pressure to produce the stoichiometric $UO_{2.0}$ sample using the humidification system are also shown in Table 4.

<Table 4>

To produce stoichiometric $UO_{2.0}$ from the unknown $UO_{2.0+x}$ sample, it was first oxidized to U_3O_8 using air gas and then reduced to $UO_{2.0}$ using a prescribed oxygen partial pressure from the possible range of pO_2 for the given temperature. Figure 3 shows the result of the calculated O/U ratio of the original sample before oxidation/reduction and is obtained from the measured sample mass change in the TGA between the equilibrium states of U_3O_8 and $UO_{2.0}$. Figure 4 shows the result of the oxygen concentration recorded by the oxygen analyzer when the experiment in Figure 3 was performed. As shown in Figure 3 and Figure 4, the humidification system provided a stable oxygen partial pressure. The application of the humidification system to the TGA can be regarded as a useful method which can be substituted for the conventional dry gas mixing.

<Figure 3>

<Figure 4>

Water condensation is a concern when this procedure is employed because condensed water is able to damage sensors of the TGA and oxygen analyzer. To prevent damage from water condensation, dry air gas is flushed through the system following a test. Indeed, high amount of water vapor leading to condensation were observed in the TGA exhaust line upstream of the oxygen analyzer. In one experiment with greater than 50% H_2O mole fraction this caused the oxygen sensor to fail.

3.2. Comparison of the predicted and measured value of pO_2

Table 5 shows a comparison of the pO_2 between the wet gas system with the humidifier and the dry gas system without the humidifier. As shown in Table 5, dry gas system has a larger difference between the predicted pO_2 value and the measured pO_2 value. All measured pO_2 values are within $\pm 5\%$ of the predicted/targeted pO_2 value at the applied temperatures when the humidification system is used. This shows that **the humidification system is more stable, accurate, and reliable than the dry gas system to be used for experiments of the oxygen partial pressure measurement for the oxide fuels.**

<Table 5>

Figure 5 shows a graph of the pO_2 differences between the wet gas system and the dry gas system. In Figure 5, a larger fluctuation of the measured pO_2 values for the dry gas system is observed and greater amounts differ from the predicted/targeted pO_2 values. While, the measured pO_2 values with the humidification system are closer to the predicted/targeted pO_2 values. This result indicates that application of the humidification system is suitable for the oxygen potential measurement and has further merit for a stable pO_2 control. The greater variation in dry gas mixing is largely due to the gas flow controllers which allow for about ± 2 ml/min variation in the set gas flow rate. This fluctuation produces a variation of approximately $\pm 5\%$ of the predicted/targeted pO_2 value.

<Figure 5>

4. Conclusions

This work has been focusing on the stability of the wet gas system because the dry gas has a larger fluctuation, and also simultaneous measurements of the oxygen partial pressure, O/U ratio, and temperature were performed using the assembled unit which contains the humidification system, TGA, oxygen analyzer and gas flow controller. This humidification system was employed as an alternative method to the conventional dry gas mixing and the pO_2 results of these two approaches were compared. Through the results from this work, it is shown that simultaneous measurement of the pO_2 and O/U ratio using the humidification system is a useful procedure and also the humidification system provides for stable and accurate oxygen partial pressure. In addition, the humidification system has benefits of easy control and flexibility for producing various oxygen partial pressures with fixed hydrogen gas flow rate. These improvements notwithstanding, the user must be vigilant to avoid issues of water condensation in the TGA when using the humidification system. This system used heating tape on TGA inlets and elevated temperature of the TGA cooling water to keep the reaction gases above the dew point. The approach demonstrated in this work was shown successful when employed for nuclear fuel research.

Acknowledgements

The authors gratefully acknowledge support under subcontract from Oak Ridge National Laboratory for the DOE Fuel Cycle R&D Program Advanced Fuels Campaign and funding for this research was provided by Oak Ridge National Laboratory (ORNL).

References

- [1] Ugajin, M. Measurements of O/U ratio and oxygen potential for UO_{2+x} ($0 \leq x \leq 0.1$). J. Nucl. Sci. Technol. 1983 March; 20:228-236.
- [2] Bolind, A. M. Control of the oxygen content of the cover gas in a molten lead-bismuth eutectic system. 2006.
- [3] Fuel Cell Technologies, Inc. Certificate of Calibration. Report of Calibration.
- [4] NBS/NRC Steam Table. Vapor Pressure of Water from 0 to 370°C. New York: Hemisphere Publishing Corp.; [Cited 1984]. Available from: <http://www.phs.d211.org/>.
- [5] FactSage. The Equilibrium Module - Regular Features. Available from: <http://www.factsage.com/>.
- [6] Cambridge Sensotec Limited. Rapidox 3100Z Oxygen Analyzer. Instruction Manual Revision 3.5. Available from: <http://www.Cambridge-Sensotec.co.uk/>.
- [7] Olander, D. R. Fundamental Aspects of Nuclear Reactor Fuel Elements. Springfield, VA: National Technical Information Center; 1976.
- [8] Blackburn, P. E. Oxygen pressures over fast breeder reactor fuel. J. Nucl. Mater. 1973; 46:244-252.
- [9] Lee, S. M. Thermodynamic and Thermochemical Investigation of Advanced TRISO Coated Particle Fuels. Columbia (SC): University of South Carolina; 2011.
- [10] Imamura, M. and Une, K. High temperature steam oxidation of UO_2 fuel pellets. J. Nucl. Mater. 1997; 247:131-137.
- [11] Dobrov, B. V. Modeling of the oxidation of uranium dioxide in a steam atmosphere. Atomic Energy 1997; 82:109-115.
- [12] Rivai, A. K. Performance of oxygen sensor in lead-bismuth at high temperature. Progress in Nuclear Energy 2008; 50:575-581.

Table 1. Experimental conditions and sample characteristics.

Oxygen partial pressure Measurement	O/U ratio	Type of sample	Gas used	Conditions used for experiments
Zirconia oxygen sensor from Oxygen Analyzer	TG/DSC analyses from TGA	Powder sample of $\text{UO}_{2\pm x}$ (100mg ~ 400mg)	5% H_2 – Ar mixture	$\text{H}_2\text{O}/\text{H}_2$ at 900°C, 1000°C, 1200°C, and 1300°C

Table 2. Table of water vapor pressure. Ref. Steam Table [4]

Temperature (°C)	Vapor pressure in bar	Vapor pressure in atm
20	0.023392	0.023086
25	0.031697	0.031283
30	0.042467	0.041912
35	0.056286	0.055550
40	0.073844	0.072879
45	0.095944	0.094689
50	0.123513	0.121898
55	0.157614	0.155553
60	0.199458	0.196850
65	0.250411	0.247136
70	0.312006	0.307926
75	0.385954	0.380907
80	0.474147	0.467947

Table 3. Calculated oxygen partial pressures in the furnace of TGA with different temperatures of the humidity bottle using the standard Gibbs free energy.

- Calculated oxygen partial pressure (atm) with 0.02% H_2 - balanced Ar								
		Water temperature in the humidity bottle						
		25°C	30°C	40°C	50°C	60°C	70°C	80°C
Furnace Temperature	800°C	3.158E-15	5.668E-15	1.714E-14	4.795E-14	1.250E-13	3.060E-13	7.066E-13
	900°C	3.218E-13	5.776E-13	1.746E-12	4.886E-12	1.274E-11	3.118E-11	7.200E-11
	1000°C	1.586E-11	2.847E-11	8.607E-11	2.408E-10	6.280E-10	1.537E-09	3.549E-09
	1100°C	4.431E-10	7.953E-10	2.405E-09	6.727E-09	1.754E-08	4.293E-08	9.914E-08
	1200°C	7.876E-09	1.414E-08	4.275E-08	1.196E-07	3.119E-07	7.631E-07	1.762E-06
	1300°C	9.711E-08	1.743E-07	5.270E-07	1.474E-06	3.845E-06	9.409E-06	2.173E-05

- Calculated oxygen partial pressure (atm) with 0.1% H2 - balanced Ar								
		Water temperature in the humidity bottle						
		25°C	30°C	40°C	50°C	60°C	70°C	80°C
Furnace Temperature	800°C	1.263E-16	2.267E-16	6.855E-16	1.918E-15	5.001E-15	1.224E-14	2.826E-14
	900°C	1.287E-14	2.310E-14	6.986E-14	1.954E-13	5.097E-13	1.247E-12	2.880E-12
	1000°C	6.344E-13	1.139E-12	3.443E-12	9.632E-12	2.512E-11	6.147E-11	1.419E-10
	1100°C	1.772E-11	3.181E-11	9.619E-11	2.691E-10	7.018E-10	1.717E-09	3.966E-09
	1200°C	3.150E-10	5.655E-10	1.710E-09	4.784E-09	1.247E-08	3.052E-08	7.049E-08
	1300°C	3.884E-09	6.972E-09	2.108E-08	5.898E-08	1.538E-07	3.764E-07	8.691E-07
- Calculated oxygen partial pressure (atm) with 5% H2 - balanced Ar								
		Water temperature in the humidity bottle						
		25°C	30°C	40°C	50°C	60°C	70°C	80°C
Furnace Temperature	800°C	5.052E-20	9.069E-20	2.742E-19	7.671E-19	2.001E-18	4.895E-18	1.130E-17
	900°C	5.149E-18	9.241E-18	2.794E-17	7.817E-17	2.039E-16	4.988E-16	1.152E-15
	1000°C	2.538E-16	4.555E-16	1.377E-15	3.853E-15	1.005E-14	2.459E-14	5.678E-14
	1100°C	7.089E-15	1.272E-14	3.847E-14	1.076E-13	2.807E-13	6.869E-13	1.586E-12
	1200°C	1.260E-13	2.262E-13	6.839E-13	1.913E-12	4.990E-12	1.221E-11	2.820E-11
	1300°C	1.554E-12	2.789E-12	8.433E-12	2.359E-11	6.152E-11	1.505E-10	3.477E-10

Table 4. Experimental results for O/U ratio and equilibrium pO₂.

Furnace Temp. (°C)	Humidifier Temp. (°C)	Sample Weight	H2O/H2	O/U ratio	Measured pO2 (atm)	$\Delta G^{\circ}_{O_2}$ (J/mol)
900	35	77.2 mg	1.0935	2.0065	1.26E-17	-379,539
1000	25	100.4 mg	0.615	2.0091	2.07E-16	-382,264
1200	25	78.0 mg	0.6152	2.0074	6.16E-13	-344,353
1300	25	369.0 mg	0.6159	2.0099	1.13E-11	-329,676

Table 5. Comparison of the pO₂ between wet gas system and dry gas system.

Method	Temperature	Humidifier	Gas flow rate (ml/min)		Predicted		Measured	
		Temperature	Ar	5% H2	pO2 (atm)	Log(pO2)	pO2 (atm)	Log(pO2)
Wet gas	900°C	35°C	110	25	6.10E-17	-16.2147	3.19E-16	-15.4962
Dry gas	900°C	N/A	110	25	3.38E-26	-25.4711	8.15E-26	-25.0888
Wet gas	1000°C	35°C	110	25	3.35E-15	-14.4750	6.17E-15	-14.2097
Dry gas	1000°C	N/A	110	25	9.07E-23	-22.0424	4.02E-24	-23.3958
Wet gas	1200°C	25°C	100	20	6.43E-13	-12.1918	6.32E-13	-12.1993
Dry gas	1200°C	N/A	100	20	1.67E-16	-15.7773	6.67E-17	-16.1759
Wet gas	1300°C	25°C	100	20	8.59E-12	-11.0660	4.37E-12	-11.3595
Dry gas	1300°C	N/A	100	20	2.18E-15	-14.6615	8.14E-15	-14.0894

Figure captions

Figure 1. Schematic of the experimental assembly unit for measurement of pO_2 and O/U ratio.

Figure 2. Details of the humidifier and TGA.

Figure 3. TGA result and O/U ratio for oxidation/reduction of UO_{2+x} using the humidification system (one of the results from Table 4).

Figure 4. Measured oxygen concentration produced by the humidification system in oxidation/reduction experiments (recorded only for the isothermal reaction in Figure 3).

Figure 5. Comparison between the predicted/targeted pO_2 and the measured pO_2 for the wet gas system and the dry gas system ($\pm 5\%$ relative error of the predicted $\log(pO_2)$).

Figure 1. Schematic of the experimental assembly unit for measurement of pO_2 and O/U ratio.

Figure 2. Details of the humidifier and TGA.

Figure 3. TGA result and O/U ratio for oxidation/reduction of UO_{2+x} using the humidification system (one of the results from Table 4).

Figure 4. Measured oxygen concentration produced by the humidification system in oxidation/reduction experiments (recorded only for the isothermal reaction in Figure 3).

Figure 5. Comparison between the predicted/targeted pO₂ and the measured pO₂ for the wet gas system and the dry gas system ($\pm 5\%$ relative error of the predicted log(pO₂)).