KNUDSEN CELL-MASS SPECTROSCOPIC STUDIES ON THE RELATIONSHIP BETWEEN MOLYBDENUM OXIDATION AND IODINE VOLATILITY IN CANDU FUEL

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ABSTRACT

High-temperature Knudsen cell-mass spectroscopic experiments were carried out to determine the relationship between molybdenum oxidation and iodine volatility in CANDU® fuel. The results show that the iodine volatility is similar in systems consisting of CsI alone, CsI / UO_2, and CsI / UO_2 / MoO_x (with molybdenum in oxidation states 0, 2 and 4). The iodine volatility is much higher, however, in the CsI / UO_2 / MoO_3 system (i.e., with molybdenum in oxidation state = 6). The results suggest that the iodine volatility in fuel increases significantly if the oxidation of the molybdenum goes beyond the MoO_2 stage.

1. INTRODUCTION

In the event of a nuclear reactor accident, radioiodine released from the fuel may have significant implications for the estimated public doses, and for equipment qualification and post-accident habitability assessments [1]. There is continuing interest in studying the volatility of iodine under different conditions so that suitable measures can be put in place to contain the iodine released from the fuel under accident conditions [2-7 and references therein].

As the yield of cesium is ten times greater than that of iodine during nuclear fission in CANDU fuel [8], it is believed that over 99.9% of iodine will exist in the form of cesium iodide (CsI). Cesium iodide has a high melting point, 621°C [9]. It is involatile outside the core and will dissolve on contact with liquid water and will be retained in the containment sump[1] under most accident conditions. Under oxidizing conditions, other compounds of cesium are thermodynamically more stable than CsI (e.g., cesium molybdates and cesium uranates) which will lead to the formation of volatile elemental iodine. Also, the formation of cesium molybdate under normal reactor operating conditions after extremely long irradiation times, which can lead to a significant amount of excess oxygen in the urania lattice, has been suggested [10]. It is,

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[1] Although most dissolved iodine in the sump will remain as CsI, a small fraction may be converted into volatile forms of iodine by the water radiolysis products.

therefore, important to understand the effects of molybdenum oxidation on the volatility of iodine present in fuel. We have carried out Knudsen cell-mass spectroscopic experiments on samples of CsI and UO_2 containing molybdenum in various oxidation states to study the relationships between molybdenum oxidation and iodine volatility in fuel.

2. EXPERIMENTAL

The cesium iodide (CsI) was obtained from Aesar Johnson Matthey Inc. (Seabrook, NH, USA). Molybdenum metal powder, 100 mesh, was obtained from Johnson Matthey (Ward Hill, MA, USA). The molybdenum dioxide (MoO_2) was obtained from Alpha Products (Danvers, MA, USA). Molybdenum trioxide samples were obtained from two suppliers: Aldrich Chemical Company Inc. and Fisher Scientific Company. The chemicals were used as obtained without further purification. The uranium dioxide (UO_2) used was obtained from CANDU fuel pellets (0.7 wt % ^{235}U). The stoichiometry of unirradiated CANDU fuel is specified as $UO_{2.0003\pm0.0002}$ [7,11]. The fuel pieces from the central part of the pellet were crushed and sieved, under an inert atmosphere, to obtain particle sizes between 25 and 125 µm. The CsI stock was kept dry in a desiccator. Molybdenum metal and/or molybdenum oxides were added to the CsI/UO_2 mixture to prepare samples for Knudsen cell experiments.

The Knudsen cell-mass spectrometer system has been described elsewhere [7,12]. The solid powders were mixed to obtain samples of desired compositions for the Knudsen cell-mass spectroscopic experiments, Table 1. The mixture of the solid powders of the desired composition (~0.4 g) was added to the weighed cell and crucible. The crucible containing the solid sample was placed in the Knudsen cell and the system was pumped overnight (for >16 h), to achieve high vacuum, before heating the sample. The cell was inductively heated and the temperature of the cell body was measured by focussing a pyrometer into a blackbody cavity on the side of the cell. Mass spectra of the gases effusing out of the Knudsen cell were recorded after the cell reached the desired temperature. At the end of the Knudsen cell experiments, the sensitivity of the mass spectrometer was determined by recording the spectra of perfluorotributylamine (FC43) through a calibrated leak [7,12].

3. RESULTS AND DISCUSSION

Samples containing molybdenum in oxidation states equal to 0, 2, 4 and 6 were investigated (see Table 1). The experiments were also performed with pure CsI (sample type A) and CsI and UO_2 mixtures (sample type B) to serve as a base line for determining the effects of molybdenum oxidation on the volatility of iodine present in the nuclear fuel. Table 2 gives a list of all the experiments carried out in this study. A few experiments were carried out at temperatures as high as ~ 2600 K (~2330°C). During the high temperature experiments, it was observed that the iodine and cesium signals disappeared in the mass spectra when the temperature of the cell reached a value of about 1100 K. However, if the experiments were continued to higher temperatures, the Cs^+ and I^+ peaks reappeared at temperatures above ~1700 K [7]. We believe that the majority of the Cs^+ and I^+ signals seen at higher temperatures (>1700 K) likely arise from the re-volatilization of the cesium and iodine species that are deposited in the Knudsen cell vacuum chamber at temperatures below 1100 K from the gases effusing out of the Knudsen cell. Therefore, the majority of the experiments were stopped at around 1100 K (Table 2) as the Cs and I signals seen at higher temperatures (>1100 K) are not exclusively from the gases effusing out of the cell.

3.1 Calculation of Vapour Pressures

The iodine and cesium ions seen in the Knudsen cell-mass spectra were I^+, Cs^+, Cs^{2+}, CsI, I^+ and Cs_2I^+ [7]. The vapour pressures of their parent species were calculated from the intensities of their peaks in the mass spectra. The pressure (p_i) of a species i in the effusing gas from the Knudsen cell is given by Equation 1:

$$p_i = I_i T / (\sigma_i \gamma_i \eta_i k_{Ag} f s) \qquad (1)$$

Where: I_i = signal intensity of the cation formed from the ionization of the species, T = temperature/K, σ_i = ionization cross-section of the precursor to the ion, γ_i = the mass dependence of the quadrupole analyzer and electron multiplier efficiency for the mass of the ion, η_i = isotopic abundance, k_{Ag} = calibration constant, derived from a calibration experiment with the silver primary standard, f = correction for the area of the orifice of the cell (relative to that in the silver experiment), and s = correction for the sensitivity of the instrument on the day of the experiment (FC43 secondary standard). The accuracy of the vapour pressures determined using Knudsen cell-mass spectroscopy depends on the chemical system being studied and the uncertainties in the values of the various terms in Equation 1, particularly the value of the ionization cross section. The ionization cross sections were obtained from the literature for Cs and I [13] and calculated for diatomic species using procedure of Kordis and Gingerich [14].

3.2 Vapour Pressures

The vapour pressures for I, Cs, I_2 and CsI species were calculated from the intensities of the peaks at the mass/charge ratios of 126.9, 132.9, 253.8 and 259.8, respectively. The vapour pressure data were analyzed to determine the relationships between the molybdenum oxidation state and the volatility of the iodine species. The iodine present as iodide ions (as in CsI) is much less volatile than the elemental form of iodine (I_2) [9]. Therefore, we compared the vapour pressures of the two *molecular* iodine species (I_2 and CsI) to determine the effects of molybdenum oxidation state on the formation of volatile iodine, Table 3. The pressures listed in Table 3 are for the temperature showing the highest vapour pressure for the molecular iodine species. The results given in this table suggest that the molecular species (containing iodine) showing significant vapour pressure is CsI in systems A, B, C, D and E, but is I_2 in system F. Also, the temperatures showing highest values for the vapour pressures of molecular iodine species are lower in system F (around 500°C) than those in systems A, B, C, D and E (around 700°C). Table 3 also gives the ratios of the vapour pressures of the molecular iodine species (I_2 and CsI) for temperatures with the highest vapour pressures for the molecular species. This ratio ($\vartheta_p = p_{I2}/p_{CsI}$) is several orders of magnitude higher in system F than in the other systems studied here.

3.3 Intensities of Molecular Species and Iodine Volatility

In order to remove the effects of uncertainties and errors in the parameters used to calculate the vapour pressure data obtained in the Knudsen cell experiments, we compared the ratios of the intensities of the molecular iodine species in the same spectra, i.e., we calculated the ratio (ϑ_I) of the intensity of I_2^+ and CsI^+ ion peaks in the mass spectra:

$$\vartheta_I = I_{(I2+)} / I_{(CsI+)} \qquad (2)$$

The ratio ϑ_I is directly proportional to ϑp, the ratio of the vapour pressures of iodine (I_2) and cesium iodide (CsI). The ratio ϑ_I is independent of the possible errors in the parameters used to calculate the vapour pressures from the Knudsen cell-mass spectroscopic results. Figure 1 shows the ratios of the intensities (ϑ_I) for all the experiments carried out in this study for the temperatures showing the highest intensities (pressures) for the molecular iodine species. The results presented in Figure 1 show that the iodine volatility, as measured by the formation of the elemental iodine (I_2), the volatile form of iodine, is similar in systems A (CsI), B (CsI/UO_2), C ($CsI/UO_2/Mo$), D ($CsI/UO_2/Mo/MoO_2$) and E ($CsI/UO_2/MoO_2$). However, the iodine volatility is much higher in system F ($CsI/UO_2/MoO_3$). The similarity of the iodine volatility in systems A, B, C, D and E suggests that the iodine volatility is not affected by the molybdenum oxidation unless it has gone beyond the MoO_2 stage. If the molybdenum oxidation has proceeded to the MoO_3 stage (system F), the iodine volatility in fuel is increased by several orders of magnitude.

There are several publications that report the results of Knudsen cell-mass spectroscopic investigations on the effects of fuel and/or molybdenum oxidation on the volatility of the iodine present in the fuel [3-7, and references therein]. However, none of these publications report the relative intensities of the molecular iodine ions (i.e., I_2^+ and CsI^+). A report by McFarlane and LeBlanc, on the Knudsen cell-mass spectroscopic experiments with a sample of CsI and U_3O_8, contains the intensity data for the I_2^+ and CsI^+ ions [4]. We calculated the ratios of the intensities for the I_2^+ and CsI^+ ions (ϑ_I), at the temperatures showing highest intensities (pressures) for the molecular iodine species from the data in their report. The results obtained from the data in Ref. 4 for the CsI and U_3O_8 system are also shown in Figure 1. The results for the CsI and U_3O_8 system are in excellent agreement with our results for the $CsI/UO_2/MoO_3$ system (system F). It is well known that iodine (I_2) is liberated on heating a sample of CsI and U_3O_8 because iodide ions (oxidation state $= -1$) are oxidized to elemental iodine (oxidation state $= 0$). The agreement between the ratios of the intensities for the I_2^+ and CsI^+ ions, in the Knudsen cell-mass spectroscopic experiments on the $CsI/UO_2/MoO_3$ system, as well as experiments on CsI and U_3O_8, supports the conclusion that the high values of this ratio (ϑ_I) indicate formation of volatile iodine. Therefore, the ratio (ϑ_I) can be used to monitor the volatility of the iodine in fuel under different conditions using Knudsen cell-mass spectroscopy.

Ugajin et al. [6] concluded that the threshold oxygen potential for the liberation of "gaseous iodine" by the reaction of CsI with UO_{2+x} (to form $Cs_2U_4O_{12}$) is -255 kJ/mol at 1073 K. An oxygen potential of –255 kJ/mol corresponds to fuel oxidation to the $UO_{2.02}$ state [15]. Therefore, it can be concluded from the results of Ugajin et al. that the oxidation of the UO_2 fuel to the $UO_{2.02}$ state *may* lead to the volatilization of the fission product (FP) iodine. Oxygen potentials of MoO_2 and MoO_3 are calculated to be -407 kJ/mole and -177 kJ/mole, respectively, at 1000 K [15]. An oxygen potential of -177 kJ/mol (i.e., equal to the oxygen potential of MoO_3) corresponds to a fuel oxidation to $UO_{2.2}$ state. Therefore, the results obtained here suggest that the oxidation of the fuel to the $UO_{2.2}$ state *will* certainly lead to the volatilization of the FP iodine.

4. SUMMARY AND CONCLUSIONS

The effects of molybdenum oxidation on iodine volatility in nuclear fuel were investigated using Knudsen cell-mass spectroscopy. Experiments were carried out with samples containing CsI and UO_2 mixed with molybdenum in oxidation states equal to 0, 2, 4 and 6. It was found that the ratio of the intensities of molecular iodine ions (I_2^+ and CsI^+) in the Knudsen cell-mass spectroscopic experiments could be used to monitor the formation of volatile iodine in fuel. The ratio (ϑ_I) can be used to determine the effects of different conditions on iodine volatility in fuel – a higher value of the ratio (ϑ_I) indicates higher volatility of iodine.

The experiments described in this report show that the iodine volatility is similar in systems consisting of CsI, $CsI+UO_{2.00}$, $CsI+UO_{2.00}+Mo$, $CsI+UO_{2\cdot00}+Mo+MoO_2$ and $CsI+UO_{2.00}+MoO_2$; however, the iodine volatility is much higher in systems consisting of $CsI+UO_2+MoO_3$. The iodine volatility is not affected until the molybdenum oxidation has proceeded beyond the MoO_2 stage. If the molybdenum oxidation proceeds to the MoO_3 stage, the iodine volatility increases by several orders of magnitude. The volatility of iodine in a sample of $CsI+UO_2+MoO_3$ is similar to that in a sample of CsI and U_3O_8.

The oxidation of the UO_2 fuel to the $UO_{2.02}$ state may lead to the volatilization of the FP iodine. Oxidation of the fuel to the $UO_{2.2}$ state will certainly lead to the volatilization of the iodine. Oxidation potential of MoO_2 is less than $UO_{2.02}$ and that of MoO_3 is equal to $UO_{2.2}$. Additional studies are required to ascertain the rate of iodine volatilization for fuel oxidation between $UO_{2.02}$ and $UO_{2.2}$.

The results presented in this report not only provide a solid experimental foundation for the thermochemical calculations that show increased iodine volatility with increasing Mo oxidation state, but also establish the extent of Mo oxidation required before it has a significant effect on iodine volatilty. This information may be used for SOURCE [16] and SOPHAEROS [17] validation, and to support iodine volatility assumptions made in CANDU safety analysis.

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TABLE 1: SYSTEMS INVESTIGATED USING KNUDSEN CELL-MASS SPECTROSCOPY

Sample	Sample Designation	Composition[a]					Molybdenum Oxidation State[b]
		CsI	UO_2	Mo	MoO_2	MoO_3	
A	CsI	1					N/A
B*	CsI/UO_2	1	24				N/A
C	CsI/ UO_2/Mo	1	24	2			0
D	CsI/ UO_2/Mo/MoO_2	1	24	1	1		+2
E	CsI/ UO_2/MoO_2	1	24		2		+4
F	CsI/ UO_2/MoO_3	1	24			2	+6

[a] Relative molar concentration.

[b] "Nominal average" oxidation state of molybdenum in the sample at the start of the experiment.

* This concentration (i.e., relative amounts of CsI and UO_2) was selected to be compatible with the previous studies carried out on the CsI/UO_2 system at AECL [7].

TABLE 2: KNUDSEN CELL-MASS SPECTROSCOPIC EXPERIMENTS WITH CESIUM IODIDE - URANIUM DIOXIDE - MOLYBDENUM SAMPLES

Sample Type #	Sample[a]	Experiment File Name	Crucible Used	Temperature Range[b] (°C)
A1	CsI	Au9918	Tantalum	373 – 211
A2	CsI	No9924	Tantalum	383 - 731
B1	CsI/UO_2	Au9927	Tantalum	376 - 2107
B2	CsI/UO_2	No9926	Tantalum	393 - 750
C1	$CsI/UO_2/Mo$	Se9915	Tantalum	380 - 2328
C2	$CsI/UO_2/Mo$	Oc9918	Tantalum	379 - 994
C3	$CsI/UO_2/Mo$	Oc9925	Tantalum	377 - 1868
C4	$CsI/UO_2/Mo$	No9930	Tantalum	376 - 1853
C5	$CsI/UO_2/Mo$	My0024	Alumina	413 - 769
D1	$CsI/UO_2/Mo/MoO_2$	Ju0026	Tantalum	409 - 721
D2	$CsI/UO_2/Mo/MoO_2$	Au0014	Tantalum	434 - 726
E1	$CsI/UO_2/MoO_2$	Ju0002	Tantalum	397 - 878
E2	$CsI/UO_2/MoO_2$	Ju0005	Tantalum	433 - 759
E3	$CsI/UO_2/MoO_2$	Ju0008	Tantalum	407 - 842
E4	$CsI/UO_2/MoO_2$	Ju0019	Tantalum	414 - 854
F1	$CsI/UO_2/MoO_3$ (A)	Se9901	Tantalum	382 - 2329
F2	$CsI/UO_2/MoO_3$ (A)	Oc9914	Tantalum	374 - 1003
F3	$CsI/UO_2/MoO_3$ (A)	Oc9927	Tantalum	376 - 1928
F4	$CsI/UO_2/MoO_3$ (A)	De9902	Tantalum	373 - 1846
F5	$CsI/UO_2/MoO_3$ (A)	My0026	Tantalum	395 - 787
F6	$CsI/UO_2/MoO_3$ (F)	De9906	Tantalum	375 - 1860
F7	$CsI/UO_2/MoO_3$ (F)	De9908	Tantalum	378 - 1900
F8	$CsI/UO_2/MoO_3$ (F)	My0031	Tantalum	401 - 732

[a] (A) and (F) stand for MoO_3 samples obtained from Aldrich and Fisher, respectively.

[b] All the experiments were carried out with the temperature in the ascending mode.

TABLE 3: VAPOUR PRESSURES OF MOLECULAR IODINE SPECIES IN THE KNUDSEN CELL-MASS SPECTROSCOPIC EXPERIMENTS[a]

Sample Type	Sample[b]	Temp. for Maximum I_2 Pressure (°C)	I_2 Pressure (atm)	Temp. for Maximum CsI Pressure (°C)	CsI Pressure[a] (atm)	Pressures Ratio[d] ϑ_p
A	CsI		Not Cal'd[c]	784	1.96E-04	
A	CsI		Not Cal'd[c]	731	1.62E-04	
B	CsI/UO_2		3.33E-10	779	6.04E-05	5.51E-06
B	CsI/UO_2		4.24E-09	697	5.57E-05	7.61E-05
C	CsI/UO_2/Mo					
C	CsI/UO_2/Mo		Not Cal'd[c]	698	1.09E-05	
C	CsI/UO_2/Mo		Not Cal'd[c]	685	2.13E-05	
C	CsI/UO_2/Mo		Not Cal'd[c]	666	1.20E-05	
C	CsI/UO_2/Mo		Not Cal'd[c]	659	8.13E-05	
D	CsI/UO_2/Mo/MoO_2		9.98E-10	706	6.61E-06	1.51E-04
D	CsI/UO_2/Mo/MoO_2		4.47E-10	683	2.51E-05	1.78E-05
E	CsI/UO_2/MoO_2		Not Cal'd[c]	703	1.51E-06	
E	CsI/UO_2/MoO_2		Not Cal'd[c]		Not Cal'd	
E	CsI/UO_2/MoO_2		Not Cal'd[c]	649	5.66E-07	
E	CsI/UO_2/MoO_2		Not Cal'd[c]	670	2.83E-06	
F	CsI/UO_2/MoO_3 (A)	487	5.47E-06		1.37E-08	3.99E+02
F	CsI/UO_2/MoO_3 (A)	495	1.86E-04		3.69E-07	5.04E+02
F	CsI/UO_2/MoO_3 (A)	524	7.04E-05		Not Cal'd	
F	CsI/UO_2/MoO_3 (F)					
F	CsI/UO_2/MoO_3 (F)	496	2.02E-05		1.16E-07	1.74E+02
F	CsI/UO_2/MoO_3 (F)	517	3.03E-05		2.39E-09	1.27E+04

[a] For temperatures showing the highest values of vapour pressures for the molecular iodine species, i.e., I_2 or CsI.

[b] (A) and (F) stand for MoO_3 from Aldrich and Fisher, respectively.

[c] Pressure not calculated because the intensity of the ion peak in the mass spectrum was very low.

[d] Ratio of vapour pressures of I_2 and CsI (i.e., p_{I2}/p_{CsI}).

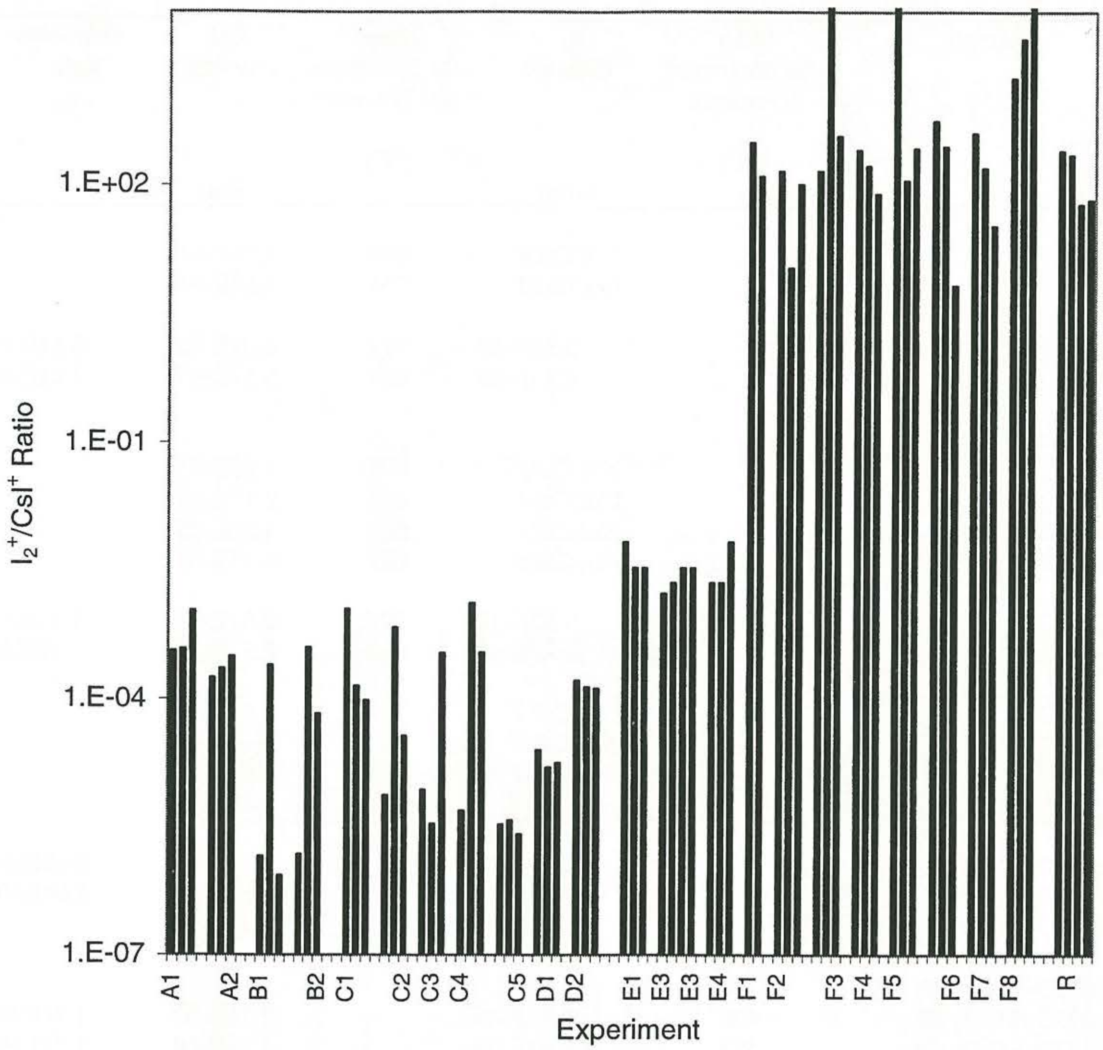

FIGURE 1: RATIO OF I_2^+/CSI$^+$ INTENSITIES (ϑ_I) IN THE KNUDSEN CELL EXPERIMENTS, AT TEMPERATURES SHOWING THE HIGHEST VALUES OF THE VAPOUR PRESSURES FOR THE MOLECULAR IODINE SPECIES FOR DIFFERENT SAMPLES CONTAINING CESIUM IODIDE. THE SYMBOLS A TO F REFER TO EXPERIMENTS OUTLINED IN TABLES 1 AND 2. R REPRESENTS A SAMPLE OF CsI+U_3O_8, DATA FROM REFERENCE 4.