OAK RIDGE NATIONAL LABORATORY

OPERATED BY

UNION CARBIDE CORPORATION

NUCLEAR DIVISION

POST OFFICE BOX X

OAK RIDGE, TENNESSEE 37830

WM Record File B-0287

WM Project 10, 11, 16

Docket No.

PDR ✓

LPDR B, N, S

March 8, 1984

Distribution:

DBROOKS

(Return to WM, 623-SS)

Dr. F. R. Cook
U.S. Nuclear Regulatory Commission
P.O. Box 1186
Richland, Washington 99352

Dear Bob:

As a result of your most recent request for information concerning the production rate of oxygen from naturally occurring radionuclides, I am enclosing a document prepared by H. C. Claiborne concerning estimates of oxygen production in the groundwater from the radiations of dissolved uranium and ^{40}K. You should note that these production rates assume that the natural uranium is in equilibrium with its daughters and that no recombination or other oxygen-consuming reaction occurs. The radiation source terms are given in the text of the document in terms of disintegrations per liter per year.

Concerning radiolysis from the radiation emitted by uranium in the basalt rock, we have determined that the average uranium content of the rock is 1.1 ppmw for the high-Mg flows and 1.5 ppmw for the low-Mg flows. We are not prepared to take this analysis further at this time due to uncertainties in the relevant geometries and radiation absorption which lead to substantial calculational complexities.

Sincerely,

Allen

Allen G. Croff, Manager
Planning & Waste Management Analysis
Chemical Technology Division

AGC:arc

Enclosure

cc: D. J. Brooks
H. C. Claiborne
A. P. Malinauskas
S. K. Whatley
R. G. Wymer
AGC File

WM DOCKET CONTROL CENTER

'84 MAR 12 P

8411080113 840308
PDR WMREB EXIORNL
B-0287 PDR

OXYGEN PRODUCTION IN BASALT GROUNDWATERS FROM DECAY OF DISSOLVED URANIUM

H. C. Claiborne

The production of hydrogen and oxygen species is governed by the G values (defined as the number of atoms or molecules of a chemical species generated by absorption of 100 e.v. of energy). The concentration or fugacity of oxygen at equilibrium is very dependent on the other species present in solution and the surrounding rock minerals that influence recombination reactions. Consequently, calculation of the oxygen fugacity would entail many assumptions in regard to the existing conditions and kinetic and thermodynamic data. To put the oxygen production rate from radiolysis from uranium decay in perspective, a comparison is made with the production rate from the decay of ^{40}K contained in basalt groundwater. Many species of oxygen are possible but for simplification, it is assumed that all oxygen is produced as O_2 molecules.

G Values

For pure water at room temperature, $G(H_2) = 1.5$ for 6 MeV alpha particles,[1] and $G(H_2) = 0.44$ for beta and gamma radiation.[2]

If all oxygen is produced as O_2,

$$G(O_2) = \tfrac{1}{2}\, G(H_2) \text{(ref. 3)} .$$

The higher temperatures in the Grande Ronde and the other dissolved species of basalt groundwater will not significantly affect these production rates.

Energy Release from Uranium and Potassium

The abundance of natural radioactive potassium (^{40}K) is 0.0118 atom per cent and it decays with a half life of 1.28×10^9 y. The decay emissions are a beta particle (89%) with a maximum energy of 1.314 MeV and a 1.460 MeV (11%) gamma photon.

Decay by beta particle emission results in a spectrum of beta particle energies up to the maximum. The difference between the maximum energy and the actual energy of the beta particle is emitted as neutrino energy. Usually the beta particle energy averages about 40% of the maximum. In the case of ^{40}K, a computer program[5] involving the theory of beta decay was used to estimate the average value, which was 38% of the maximum. Therefore, decay results in a ($\beta + \gamma$) energy release of

$$0.38 \times 1.314 \times 0.89 + 1.460 \times 0.11 = 0.606 \text{ MeV} .$$

Natural uranium forms two decay series - the uranium (radium) series starting with ^{235}U. Table 1 shows the uranium series[6] with the emission characteristics for the parent and each daughter; Table 2 shows the same for the actinium series.[7]

Assuming secular equilibrium, each decay of a parent is matched by a daughter decay and total energy release is obtained by summing the energy release for each nuclide with each particle or photon being weighted by its abundance. Since the contribution from the β energy is relatively small; for simplication it was assumed that the average β energy was 40% of the maximum.

For the uranium series:

Total α energy = 42.82 MeV

Maximum β energy = 5.84 MeV

γ energy = 1.92 MeV

Total (β_{av} + γ) energy = 4.22 MeV .

For the actinium series:

Total α energy = 36.97 MeV

Maximum β energy = 2.97 MeV

γ energy = 0.47 MeV

Total (β_{av} + γ) energy = 1.66 MeV

In the following calculations, it is assumed that each decay of U^{238} and U^{235} produces the energies listed above and that natural uranium is composed of 99.29% ^{238}U and 0.71% ^{235}U.

Uranium and Potassium Concentrations

The Middle Sentinel Bluffs and Umtanum flows are the candidate repository horizons. The site characterization report[8] lists the following concentration ranges for the potassium content of their groundwater:

Middle Sentinel Bluffs: 12 to 16 mg/L; average 14 mg/L

Umtanum: 3.3 to 8.1 mg/L; average 5.9 mg/L.

The BWIP estimated[9] the solubility of uranium synthetic groundwater to be in the range of 1.3×10^{-11} to 1.9 to 10^{-9} moles/L with an expected value of 1.0×10^{-10} moles/L. A conservative value, which is the highest found in any experiment, is listed as 1.0×10^{-6} moles/L. These values are 3.2×10^{-6}, 4.5×10^{-4}, 2.4×10^{-5}, and 0.24 mg/L or ppm, respectively.

Calculations

^{40}K contribution for 16 mg/L or ppm:

$(16 \times 10^{-3})(1.18 \times 10^{-4})(6.02 \times 10^{23})/39.1 = 2.90 \times 10^{16}$ atoms/L

$(2.90 \times 10^{16})(0.6931)/1.28 \times 10^{9} = 1.57 \times 10^{7}$ dis./L-yr.

$(1.57 \times 10^{7})(0.606 \times 10^{6})(0.22)/100 = 2.10 \times 10^{10}$ O_2 moles/L-yr.

^{238}U contribution for 1.0×10^{-6} moles U/L:

$(10^{-6})(0.9929)(6.02 \times 10^{23})(0.6931)/4.49 \times 10^{9} = 9.22 \times 10^{7}$ dis./L-yr.

From $(\beta_{aw} + \gamma)$, $(9.22 \times 10^{7})(10^{6})(4.22)(0.22)/100 = 8.57 \times 10^{11}$ O_2 moles/L-yr.

From α, $(9.22 \times 10^{7})(10^{6})(42.82)(0.75)/100 = 2.96 \times 10^{13}$ O_2 moles/L-yr.

^{235}U contribution for 1.0×10^{-6} moles U/L:

$(10^{-6})(0.0071)(6.02 \times 10^{23})(0.6931)/7.10 \times 10^{8} = 4.17 \times 10^{6}$ dis./L-yr.

From $(\beta_{aw} + \gamma)$, $(4.17 \cdot 10^{6})(1.66)(10^{6})(0.22)/100 \doteq 1.52 \times 10^{10}$ O_2 moles/L-yr.

From α, $(4.17 \times 10^{6})(36.97)(10^{6})(0.75)/100 = 1.16 \times 10^{12}$ O_2 moles/L-yr.

Total for uranium $= 3.17 \times 10^{13}$ O_2 moles/L-yr.

The O_2 production rates for other concentrations can be obtained by multiplying these results by the ratios of the concentrations.

Calculated Results

The O_2 production rates for the range of concentrations listed previously are given in the following table.

	Uranium		^{40}K, Mid. Sent. Bluff		^{40}K, Umtanum	
	Conc., ppm	O_2 prod., moles/L-yr	conc, ppm	O_2 prod., moles/L-yr	conc, ppm	O_2 prod., moles/L-yr
high	4.5 - 4*	6.0 + 10	16	2.1 + 10	8.1	1.1 + 10
expected or average	2.4 - 5	3.2 + 9	14	1.8 + 10	5.9	7.7 + 9
low	3.1 - 6	4.1 + 8	12	1.6 + 10	3.3	4.3 + 9
conservative	2.4 - 1	3.2 + 13	-	-	-	-

*Read as 4.5×10^{-4}.

Discussion

The values for the uranium concentrations were based on solubilities for conditions around the waste package. The conservative value of 240 ppb for uranium is very high (conditions of experiment that obtained this value are unknown).

If such a high concentration occurs, it cannot be expected to be sustained once the groundwater leaves the repository region and interacts with surrounding groundwater. Consequently, the expected value for the uranium concentration seems more likely. The Cohassett flow of the Middle

Sentinel Bluffs sequence is now the most favored horizon for the repository. Therefore, the ^{40}K and natural uranium contained in the region groundwater can be expected to exert the greatest influence on the oxygen production by radiolysis of the water; i.e., leached uranium should not significantly affect the oxygen fugacity due to oxygen production in the far-field and probably not in the near-field.

Table 1. Uranium (radium) series

Isotope	Symbol	Half-life	Radiation	Energy[a] (MeV)
Uranium-238	^{238}U	4.5×10^9 y	α	4.18(77), 4.13(23)
Thorium-234	^{234}Th	24.1 d	β	0.19(65), 0.10(35)
			γ	0.09(15), 0.06(7), 0.03(7)
Protactinium-234	^{234}Pa	1.18 min.	β	2.31(93), 1.45(6), 0.55(1)
			γ	1.01(2), 0.77(1), 0.04(3)
Uranium-234	^{234}U	2.50×10^5 y	α	4.77(72), 4.72(28)
			γ	0.05(28)
Thorium-230	^{230}Th	8.0×10^4 y	α	4.68(76), 4.62(24)
Radium-226	^{226}Ra	1622 y	α	4.78(94), 4.59(6)
			γ	0.19(4)
Radon-222	^{222}Rn	3.82 d	α	5.48(100)
Polonium-218	^{218}Po	3.05 min.	α	6.00(100)
Lead-214	^{214}Pb	26.8 min.	β	1.03(6), 0.66(40), 0.46 (50), 0.40(4)
			γ	0.35(44), 0.29(24), 0.24 (11), 0.05(2)
Bismuth-214	^{214}Bi	19.7 min.	β	3.18(15), 2.56(4), 1.79(8), 1.33(33), 1.03(22), 0.74(20)
			γ	2.43(2), 2.20(6), 2.12(1), 1.85(3), 1.76(19), 1.73(2), 1.51(3), 1.42(4), 1.38(7), 1.28(2), 1.24(7), 1.16(2), 1.12(20), 0.94(5), 0.81(2), 0.77(7), 0.61(45)
Polonium-214	^{214}Po	160×10^{-6} s	α	7.68(100)
Lead-210	^{210}Pb	19.4 y	β	0.06(17), 0.02(83)
			γ	0.05(4)
Bismuth-210	^{210}Bi	5.0 d	β	1.16(100)
Polonium-210	^{210}Po	138.4 d	α	5.30(100)
Lead-206	^{206}Pb	Stable		

[a]Numbers in parentheses indicate percent abundance.

Taken from Reference 4.

Table 2. Actinium series

Isotope	Symbol	Half-life	Radiation	Energy[a] (MeV)
Uranium-235	^{235}U	7.1×10^{8} y	α	4.40(57), 4.37(18), 4.58(8)
			γ	0.18(54), 0.14(11), 0.20(5)
Thorium-231	^{231}Th	25.5 h	β	0.14(45), 0.30(40), 0.22(15)
			γ	0.08(10), 0.03(2)
Protactinium-231	^{231}Pa	3.25×10^{4} y	α	5.01(24), 5.02(23), 4.95(22)
			γ	0.29(6), 0.03(6)
Actinium-227	^{227}Ac	21.6 y	α	4.95(1.2), 4.86(0.18)
			β	0.043(99+)
			γ	0.070(0.08)
Thorium-227	^{227}Th	18.2 d	α	5.98(24), 6.04(23), 5.76(21)
			γ	0.24(15), 0.31(8), 0.050(8)
Radium-223	^{223}Ra	11.43 d	α	5.71(54), 5.61(26), 5.75(9)
			γ	0.27(10), 0.15(10), 0.33(6)
Radon-219	^{219}Rn	4.0 s	α	6.82(81), 6.55(11), 6.42(8)
			γ	0.27(9), 0.40(5)
Polonium-215	^{215}Po	1.78×10^{-3} s	α	7.38(100)
Lead-211	^{211}Pb	36.1 min.	β	1.39(88), 0.56(9), 0.29(1.4)
			γ	0.83(3.4), 0.40(3.4), 0.43(1.8)
Bismuth-211	^{211}Bi	2.15 min.	α	6.62(84), 6.28(16)
			γ	0.35(14)
Thallium-207	^{207}Tl	4.79 min.	β	1.44(99.8)
			γ	0.90 (0.16)
Lead-207	^{207}Pb	Stable		

[a]Numbers in parentheses indicate percent abundance.

Taken from Reference 4.

References

1. G. H. Jenks, Radiolysis and Hydrolysis in Salt-Mine-Brines, p. 51, ORNL/TM-3717 (March 1972).

2. G. H. Jenks, Radiation Chemistry of Salt-Mine Brines and Hydrates, p. 38, ORNL-5726 (July 1981).

3. G. H. Jenks, Effects of Gaseous Radioactive Nuclides on the Design and Operation of Repositories for Spent LWR Fuel in Rock Salt, p. 87, ORNL-5578 (December 1979).

4. U.S. Environmental Protection Agency, Radiological Quantity of the Environment, pp. 26-27, EPA-520/1-76-010 (May 1976).

5. N. B. Gove and M. J. Martin, Nuclear Data Tables A10; Log-f. Tables for Beta Decay (1971).

6. Op. Cit., U.S. Environmental Protection Agency, p. 30.

7. Ibid., p. 32.

8. Rockwell Hanford Operations, Site Characterization Report, DOE/RL 82-3, Vol. II, p. 6.2-3 (November 1982).

9. Rockwell Hanford Operations, BWIP Data Package for Reference Solubility and Kd Values, SD-BWI-DP-001, p. 7 (June 1983).