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BEHAVIOR OF CARBON-14 IN WASTE PACKAGES FOR SPENT FUEL IN A REPOSITORY IN TUFF

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ABSTRACT

Analysis of gas from a heated air-filled canister containing a spent fuel assembly before and after rupture of a fuel rod shows that about 1.5 mCi of ^{14}C from the external surface of the assembly was rapidly oxidized and released as $^{14}CO_2$ in excess oxygen at 275°C and 10^4 rad/hr. After rupture, an additional 0.3 mCi was released, probably also from the external surface. The total ^{14}C inventory in the entire 15x15 rod assembly including structural hardware is estimated to be 690 mCi. These measurements indicate that account will have to be taken of the time distribution of lifetimes of the canisters, and a broad definition of the "engineered system" may be necessary, in order to meet 10CFR60 requirements with spent fuel in a repository in tuff.

INTRODUCTION

Under the direction of the Office of Civilian Radioactive Waste Management, the Department of Energy's Nevada Nuclear Waste Storage Investigations (NNWSI) project is evaluating a candidate site in tuff rock at Yucca Mountain, Nevada, for potential location of a high level nuclear waste repository. Lawrence Livermore National Laboratory, a participant in the NNWSI project, is responsible for design and performance analysis of waste packages for this site. NNWSI proposes to locate the repository above the water table in the unsaturated zone. In such an environment, it is possible that volatile radionuclides could move by vapor phase transport. Accordingly, the Nuclear Regulatory Commission (NRC) has proposed amendments [1] to federal regulation 10CFR60 [2] to include consideration of vapor transport of radionuclides in candidate repositories in the unsaturated zone. Of particular concern are species that have sufficiently long half-lives that they will have significant remaining activities after the 300 to 1000-year containment period specified in 10CFR60. Spent reactor fuel is currently considered to be the most probable commercial high level nuclear wasteform in the U.S. The long-lived radionuclides that could enter the vapor phase at spent fuel storage temperatures are ^{14}C (as, for example, CO_2, CO, or CH_4) and ^{129}I. NRC regulation 10CFR60 limits the release rate of any radionuclide from the engineered barrier system following the containment period to one part in 100,000 per year of the inventory present at 1,000 years after permanent closure of the repository. This requirement does not apply to any radionuclide which is released at a rate less than 0.1% of the calculated total release rate limit, which is one part in 100,000 per year of the total inventory of radioactive waste remaining after 1000 years.

In addition to the NRC regulation, the Environmental Protection Agency (EPA) has proposed regulation 40CFR191 [3] which would limit the cumulative release of ^{14}C to the accessible environment to 0.1 curie per MTHM (metric tonne of heavy metal) for a period of 10,000 years after disposal.

In order to design a waste package that will meet these limitations on ^{14}C release, it is necessary to have several pieces of data. Two of the most important are the inventory of ^{14}C in spent fuel and the fraction of this inventory that will be in the gaseous state or will readily enter it when the package and fuel cladding are eventually breached. The ^{14}C inventory in spent fuel has been calculated by a number of workers, and recent reviews have been given by Braun et al. [4] and by Bush [5]. The most comprehensive calculations for U.S. spent fuel are probably those of Croff and Alexander [6], building upon the work of Davis [7]. These workers have computed ^{14}C inventories of 1.62 and 1.55 Ci/MTU for BWR and PWR spent fuel respectively, of which 1.08 and 0.95 Ci/MTU, respectively, are in the structural parts. The burnups were taken to be 27,500 and 33,000 MWd/MTU, respectively. The main difficulty involved in performing such calculations is to obtain reliable values for the N impurity concentrations in the components of the fuel assemblies when they were new, since the largest contribution of ^{14}C comes from ^{14}N (n,p) ^{14}C reactions. $^{17}O(n,\alpha)^{14}C$ reactions and ternary fission also contribute. Crow [8] reported that the N concentrations in UO_2 from five fabrication plants ranged from 1 to 100 ppm. Davis found values ranging from <10 to >50 ppm, with average single plant values from 2.8 to 47.8 ppm. Barner [22] reported measurements of ^{14}C concentrations in fuel and cladding from the H. B. Robinson PWR. (Note that the numbers printed in Ref. 22 are erroneously a factor of 1000 smaller than the actual measured values, according to Barner.) For a measured burn-up of 31,660 MWd/MTU, the results were 3.30×10^{-7} and 6.00×10^{-7} Ci/g (sample) for fuel and cladding, respectively. These numbers translate to 0.37 and 0.16 Ci/MTU, respectively. Measurements have been reported of gaseous ^{14}C releases from fuel reprocessing plants [9], and these releases appear to represent a substantial fraction of calculated values for the UO_2 component of spent fuel assemblies. In addition to the ^{14}C produced within the fuel assemblies themselves, a substantial amount is also produced in the reactor cooling water from oxygen and the small amounts of dissolved nitrogen. Although much of this is released in the off-gas stream from operating reactors, some may be deposited onto the external surfaces of the fuel assemblies.

With regard to the fraction of the ^{14}C in spent fuel that is in the gaseous state or is readily available to enter it upon oxidation, even less is known. It is difficult to calculate this parameter because of the complexity of the chemical system involved, and we are not aware of any published measurements. Fortunately an opportunity to perform some measurements has arisen in the course of other work that was underway at our Laboratory in support of spent fuel storage tests at the Nevada Test Site. These tests [10] are being performed by Pacific Northwest Laboratory in cooperation with the Westinghouse Waste Technology Services Division. Our laboratory was asked to perform mass spectrometric and radiometric analyses of gas samples taken from spent fuel storage canisters in order to determine whether any fuel rods had ruptured. The species of primary interest as an indicator of cladding breach was ^{85}Kr. When the analyses also revealed ^{14}C, we decided to pursue the work further in support of the high level waste package task. This paper discusses the initial results obtained and the ramifications for the storage of spent fuel in a potential repository in tuff.

EXPERIMENTAL MATERIALS, EQUIPMENT, AND PROCEDURE

The spent fuel assembly [11] used in the Fuel Temperature Test was manufactured by Westinghouse prior to 1972. It is a 15x15-rod pressurized water reactor assembly. It consists of UO_2 fuel pellets inside Zircaloy-4 tubes, held in place by type 304 stainless steel orifice plates and Inconel 718 spacer grids. The concentrations of nitrogen impurity in the UO_2 and structural parts are either not known or are considered proprietary by the manufacturer. Before the tubes were welded shut, they were filled with helium gas to a pressure of about 2.31 MPa (335 psig) without removing the residual air. The purity of the helium gas is considered proprietary by the manufacturer. The assembly measures 0.21x0.21x4.10 m, weighs 0.645 Mg, and was originally fueled with 0.448 MT of 2.559 wt.% ^{235}U-enriched uranium.

The assembly is designated B02, and was used in the Turkey Point Unit 3 reactor, owned and operated by Florida Power and Light Co. for the first two cycles of operation, extending from January 12, 1972 to November 25, 1975. The calculated burnup for this assembly was given as 25,665 MWd/MTU [10,11]. Measured burnups for the companion assembly B17 ranged from 19,890 to 27,700 MWd/MTU [15].

After removal from the reactor the assembly was stored in a water pool for less than 2 years. It was then shipped under dry conditions to Battelle Columbus Laboratories, where it underwent various non-destructive measurements and examinations in air and water environments. Then it was shipped to the Nevada Test Site [12], arriving on December 4, 1978. Two days later it was sealed inside a helium-filled canister, and was used in a silo-type fuel storage test for about 4 years. The assembly was removed from its canister and inspected, and two fuel rods were removed. The assembly was then placed in the Fuel Temperature Test (FTT) stand.

The FTT stand incorporates a stainless steel canister fitted with a bolted closure lid from which 15 thermowells are suspended. The wells fit inside the fuel assembly guide thimbles and contain thermocouples. The closure lid contains a fitting which connects to an atmosphere control system. Electrical heaters are mounted on a liner which surrounds the canister and are used to control the temperature of the assembly.

In the test that was performed [13], the canister was filled with air and the temperature was raised to 275°C and was programmed to decrease slowly with time. Gas samples were taken at the beginning and periodically thereafter to determine whether any cladding breaches had occurred. Canister temperature and pressure were monitored.

Gas samples [10] were taken by Westinghouse using 500-mL stainless steel sampling bottles onto each of which a shut-off valve had been welded. Before use, each bottle was baked out at about 200°C to a pressure of about 10^{-5} Pa (about 10^{-7} torr). Samples were taken through a 15-meter long stainless steel line connected to the fitting on the canister lid. Before analytical samples were taken, the line was purged by drawing a preliminary sample which was later discarded. The vacuum on each analytical sample bottle was verified before a sample was taken. After sealing, the sample bottles were transported to our laboratory.

Mass spectrometry was performed using the CEC 21-103C gas analytical mass spectrometer in the Analytical Chemistry Section of our laboratory. This instrument is fully automated, using a Digital Equipment Corp. LSI-11.2 computer. Calibration is accomplished using pure gases and known gas mixtures. Output is analyzed by a regression-fit procedure on an LSI-11 computer.

Isolation of the various radioactive gases was accomplished by temperature-controlled elution chromatography from activated charcoal and molecular sieve (5A) columns. Known quantities of each desired constituent gas were mixed with the sample to serve as carriers prior to separation. Purity of the recovered gases was checked by mass spectrometry. Isolated ^{14}CO and $^{14}CH_4$ were converted to $^{14}CO_2$ over CuO at 480°C and 850°C respectively. The converted gases as well as the original $^{14}CO_2$ fraction were measured for radioactivity content using standard internal proportional counting techniques. ^{85}Kr, as recovered in the krypton fraction from the original separation, was also assayed by internal proportional counting in the low-level samples and by thin-window beta counting in the high-level samples. System blanks and detector background checks were conducted prior to sample processing and analyses to preclude cross-contamination between samples. Concentrations of the various constituents was computed from the measured radioactivity using known detector fill, carrier and sample volumes.

Table 1. Results of Mass Spectrometric and Radiometric Analyses of Canister Gas Samples from Fuel Temperature Test

LLNL Sample No.	Date Taken	Canister Temp. (°C)	Canister Pressure (mm Hg) Before	Canister Pressure (mm Hg) After	Mass Spectrometric Analysis (vol.%) N_2	O_2	Ar	He	CO_2	CO	N_2O	H_2O (Min)	$\frac{^{13}CO_2}{^{12}CO_2}$ (%)	Radiometric Analysis $\left(\frac{pCi}{cm^3\ STP}\right)$ ^{85}Kr	$^{14}CO_2$	^{14}CO	$^{14}CH_4$
9	3/01/83	118.9	668.0	592.4	78.4	20.6	0.93	N.D.	0.065	tr	N.D.	0.05	1.1	≤0.05	6.0	N.M.	N.M.
17	4/08/83	274.5	929.4	785.5	78.9	18.2	0.94	N.D.	1.96	N.D.	0.03	0.23	3.6	0.52	8,170	N.M.	N.M.
18	5/12/83	270.0	780.4	659.0	79.0	17.5	0.94	0.27	2.22	N.D.	0.05	0.30	4.0	122,000	10,200	N.M.	N.M.
19	6/03/83	268.0	655.7	560.6	79.4	16.9	0.96	0.32	2.31	N.D.	0.08	0.32	2.9	147,000	9,740	0.35	0.21
26	7/18/83	261.7	553.9	470.7	79.6	16.7	0.94	0.32	2.25	N.D.	0.14	0.45	3.6	147,100	11,300	N.M.	N.M.
34	8/17/83	258.6	468.7	400.4	79.7	16.5	0.95	0.34	2.33	N.D.	0.18	0.36	3.3	157,000	7,040	N.M.	N.M.
35	9/20/83	256.2	398.0	340.4	79.9	16.1	0.95	0.34	2.35	N.D.	0.23	0.53	3.5	160,800	N.M.	N.M.	N.M.
Standard Dry Atmosphere		15.0	760		78.09	20.95	0.93	5E-4	0.03	0	5E-5	0	1.12				

tr = trace, N.D. = not detected, N.M. = not measured

Table 2. Calculated Gas Volumes and Releases of ^{85}Kr and ^{14}C to Canister Gas in Fuel Temperature Test

LLNL Sample No.	Canister Gas Volume (cm^3 @ STP) Before	Canister Gas Volume (cm^3 @ STP) After	Gas Removed (cm^3 @ STP)	Air Added (cm^3 @ STP)	^{85}Kr in Canister Gas (Ci) Before	^{85}Kr in Canister Gas (Ci) After	^{85}Kr Removed in Sampling (Ci)	Apparent Cumulative ^{85}Kr Release (Ci)	^{14}C in Canister Gas (Ci) Before	^{14}C in Canister Gas (Ci) After	^{14}C Removed in Sampling (Ci)	Apparent Cumulative ^{14}C Release (Ci)
9	1.837E+5	1.629E+5	2.08E+4	0	≤9E-9	≤8E-9	≤1E-9	≤9E-9	1.10E-6	9.77E-7	1.2 E-7	1.10E-6
17	1.830E+5	1.546E+5	2.84E+4	0	9.5E-8	8.0E-8	1.5E-8	9.5E-8	1.495E-3	1.263E-3	2.32E-4	1.50E-3
18	1.549E+5	1.308E+5	2.41E+4	0	1.890E-2	1.596E-2	2.94E-3	1.89E-2	1.580E-3	1.334E-3	2.46E-4	1.81E-3
19	1.306E+5	1.117E+5	1.89E+4	0	1.920E-2	1.642E-2	2.78E-3	2.21E-2	1.272E-3	1.088E-3	1.84E-4	1.75E-3
26	1.117E+5	9.489E+4	1.68E+4	0	1.643E-2	1.396E-2	2.47E-3	2.22E-2	1.262E-3	1.072E-3	1.90E-4	1.92E-3
34	9.504E+4	8.119E+4	1.38E+4	0	1.492E-2	1.275E-2	2.17E-3	2.31E-2	6.691E-4	5.716E-4	9.75E-5	1.52E-3
35	8.107E+4	6.933E+4	1.17E+4	6.90E+4	1.304E-2	1.115E-2	1.89E-3	2.34E-2				

RESULTS

The results are shown in Table 1. Sample #9 was taken during warm-up after sealing the canister. The $^{14}CO_2$ analyses for samples #26 and #34 are somewhat suspect because of air leaks into the samples during storage prior to analysis. In these cases, the $^{14}CO_2$ concentrations were corrected by scaling by the ratio of He present in the original to that in the stored samples.

DISCUSSION

As can be seen, the initial sample (#9) has a composition similar to that of the standard atmosphere, except for a slightly elevated CO_2 level and a ^{14}C concentration well above background. The second sample (#17) shows a decrease in O_2 and an increase in CO_2, indicating that some carbon has oxidized in the presence of the excess oxygen, elevated temperature, and radiation field of about 10^4 rad/hr. Although there is an apparent increase in H_2O, these data are uncertain, because H_2O readily adsorbs on surfaces. In view of the observed high values for $^{13}C/^{12}C$ and $^{14}CO_2$ and the knowledge that the test canister was previously heated and outgassed, it seems clear that at least a substantial fraction of this carbon originated from the spent fuel assembly. Since the He and ^{85}Kr levels have not yet risen substantially, it also seems clear that the observed ^{14}C came from the external surfaces of the assembly rather than the insides of the fuel rods.

N_2O begins to be detectable in sample #17 also, and its concentration increases with time. This gas is known to be produced by the irradiation of air, and the production rate here is consistent with yields determined in other work [14]. The absence of other nitrogen species such as HNO_3 or NO_2 which are commonly seen in irradiated moist or dry air is probably related to the formation of CO_2, as has been seen before [14].

By the time sample #18 was taken, it can be seen clearly that a cladding breach has occurred, since He has appeared, and the ^{85}Kr level has greatly increased. The fraction of ^{85}Kr eventually released can be calculated by assuming that one rod was breached and applying the ideal gas law, using the measured temperatures and pressures. The canister void volume was determined to be 3.00×10^5 cm^3 (STP) from physical dimensions and by He dilution. See Table 2. Measurements by others [15] on companion fuel rods have indicated the total He gas in a single fuel rod to be 519 cm^3 STP on the average. The total ^{85}Kr released is found to be about 22 mCi, which is about 0.21% of the inventory per fuel rod calculated by scaling the results of Croff and Alexander [6] for burnup. This release fraction is in good agreement with measurements by others on companion fuel (0.3% maximum) [15].

It can be seen that both the mass spectrometric and the radiometric analyses show that CO_2 is the dominant form of C present in the gas. This is to be expected, since both CO and CH_4 are oxidized by excess O_2 in a radiation field [16].

The most important results from the standpoint of ^{14}C behavior in a repository are that considerable ^{14}C entered the gas phase before the rod breach occurred, and a smaller amount was added after the breach. As mentioned above, the ^{14}C which was observed prior to the breach had to have come from the surfaces of the fuel assembly. It could have originated from reactor transmutation of ^{14}N impurity in the Zircaloy or from reactions in the cooling water. The total amount of ^{14}C in the canister gas can also be calculated, and is shown in Table 2. At the time sample #17 was taken, it amounts to about 1.5 mCi. Assuming an initial N impurity level of 80 ppm in the Zircaloy [17], a thermal neutron flux of 2.6×10^{13} $^{n}/$cm^{2}-sec [18] and a spectrum-averaged cross section of 1.48 barns [7], one finds that the amount of released ^{14}C is equivalent to that produced in about 1% of the cladding thickness. It is interesting to note that this is approximately the average thickness of the oxide layer observed on the Zircaloy of companion rods [15]. Alternatively, various estimates [5,19] of ^{14}C production in the reactor cooling water range from about 3 to 15 Ci/GW(e)-yr, which translates to between 30 and 150 mCi for the water associated with assembly B02. Even if only a small fraction of this ^{14}C adhered to the fuel assembly, the observed release could be accounted for from this source alone.

It is not possible to determine precisely how much additional ^{14}C was added to the canister gas by the breach, because it is not known whether or not the external surfaces are still releasing ^{14}C after the breach. Also, the ^{14}C may be exchanging with a reservoir of natural carbon on the canister walls or on the walls of the sampling bottles. It should be noted that previous mass spectrometric analyses of fuel rod gas taken directly from zircaloy-clad UO_2 spent fuel assemblies [15, 20-22] have generally shown CO, CO_2, and CH_4 concentrations to be near or below the detection limits. While this suggests that little gaseous carbon is available in the fuel rod gas, it does not rule out a possible significant contribution of gaseous ^{14}C here, because of the greater sensitivity of radiometric measurements. It remains to be seen from analyses of samples now on hand whether significantly more ^{14}C was oxidized and released after oxygen could enter the breached rod.

In summary, the calculated total ^{14}C inventory in assembly B02 is 690 mCi, based on Croff and Alexander [6], adjusting for burnup. Of this, about 1.5 mCi was rapidly oxidized and released from the external surface by the heat, radiation, and excess oxygen. After a single rod from the total of 204 fuel rods ruptured, an additional 0.3 mCi was released to the canister gas. Assuming that all of this ^{14}C came from the external surface, this suggests that less than 0.3% of the ^{14}C inventory in a stored canister of spent fuel might be released as gas when the canister is breached and air is allowed to enter. It is unlikely that significantly more ^{14}C will be released as gas immediately after fuel rod cladding rupture, but direct radiometric analysis for ^{14}C should be performed on fuel rod gas to check this.

A comparison of this value with the 10CFR60 release limit of 10^{-5} per year from the "engineered system" shows that it will be important to carefully define the "engineered system" and to take account of the time distribution of lifetimes of the canisters to meet this requirement. (Of course, if these lifetimes approached 90,000 years, the ^{14}C would have decayed to below the release limit, but this would be difficult to guarantee).

In the case of 40CFR191, the limit of 0.1 Ci/MTU would not be exceeded by this immediate gaseous release. Further effort will be necessary to account for ^{14}C release to the accessible environment during the 10,000-year period as a result of slower releases due to oxidation of the UO_2 and metal parts and the transport of $^{14}CO_2$ through the geological environment.

CONCLUSIONS AND RECOMMENDATIONS

1. About 1.5 mCi of ^{14}C was rapidly oxidized and released as gas from the external surface of a PWR spent fuel assembly stored in air at 275°C and producing a radiation field of about 10^4 rad/hr.

2. An additional 0.3 mCi was released as gas after one of the 204 fuel rods breached, but it is likely that most of this also came from the external surface of the assembly. Radiometric ^{14}C analysis should be performed directly on fuel rod gas to check this conclusion.

3. This initial gaseous release would not exceed the limit of 40CFR191. Further study is needed on slower releases of ^{14}C.

4. This gaseous release is large enough that a broad definition of the "engineered system" in 10CFR60 may be necessary, and account will have to be taken of the time distribution of lifetimes of the canisters, in order to meet the requirement of this regulation.

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