U.S. DEPARTMENT OF ENERGY
OFFICE OF CIVILIAN RADIOACTIVE WASTE MANAGEMENT

NUCLEAR WASTE TECHNICAL REVIEW BOARD
FULL BOARD MEETING

SUBJECT: CARBON-14 RELEASES

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Carbon-14 (^{14}C) Production in Light-Water Reactors

Reaction	Isotopic Abundance of Reactant Nuclide	Thermal Neutron Cross Section (barns)
$^{14}N(n,p)^{14}C$	99.63%	1.8
$^{17}O(n,\alpha)^{14}C$	0.038%	0.24
$^{13}C(n,\gamma)^{14}C$	1.11%	0.001

- In light-water reactors (LWRs) the first reaction predominates in the fuel, cladding, and structural metal, because of impurity nitrogen
- The second reaction predominates in the reactor cooling water
- The third reaction is not significant in LWRs

Carbon-14 (^{14}C) Production in Light-Water Reactors

(Continued)

- The ^{14}C produced in the cooling water is mostly exhausted to the atmosphere at levels of about 10 curies per year for a 1000-MWe plant (NCRP, 1985). A small fraction goes into ion exchange resins and is disposed of as low-level waste (Hesbol, 1990)

- The ^{14}C produced in the fuel is mostly retained in the spent fuel, although some is lost from the external surface by dissolution in the water of spent-fuel pools, exchange with ventilation air, and exhausting from the stack to the atmosphere

- Reactor structural material not associated with fuel may end up as low-level waste or "greater than class C" waste

Previous Analysis of Carbon-14 Inventory in LWR Spent Fuel

- Determined by calculations and measurements
- Calculations require knowledge of average nitrogen impurity content of UO_2, zircaloy, and structural metals
- Most comprehensive calculation in U.S. was done by Wallace Davis at Oak Ridge National Laboratory (ORNL) in 1977. Subsequent work done by ORNL has used his data
- The value for UO_2 was determined by averaging measured data from reactor fuel manufacturers
- The values for the metals were estimated, making use of maximum values in American Society for Testing and Materials (ASTM) standards
- The result obtained by ORNL for the ^{14}C inventory in PWR spent fuel with 33,000 MWd/MTU burnup was 1.55 curies per metric ton of initial uranium (DOE/RW-0184, 1987)

Recently Revised Calculations of Carbon-14 Inventory in Spent Fuel

- Nitrogen impurity contents were re-examined by Van Konynenburg of LLNL
- The value for UO_2 (25Mg/gu) was retained, since it was based on measurements
- The values for the metals were revised downward to represent production averages

	N content (wppm)	
Material	ORNL value	revised value
Zircaloy	80	40 (Schemel, 1989)
304 stainless steel	1300	400 (Hickey, 1973)
Nickel alloys	1300	120 (Crum, 1989)

- The values for zircaloy and nickel alloys were obtained from discussions with producers. The value for stainless steel was obtained from the Aerospace Structural Metals Handbook
- The revised result for the ^{14}C inventory of PWR spent fuel with 33,000 MWd/MTU burnup is 1.00 curies per metric ton of initial uranium compared to the ORNL value of 1.55. (This incorporates new measurements by A.T. Luksic of PNL giving the variation of neutron flux and spectrum-averaged cross section in LWR cores)

Measurements of Carbon-14 in LWR Spent Fuel

- Direct measurements have been made (mostly by the Materials Characterization Center at PNL) for only a few fuel assemblies
- Measurements on all the components of an assembly have been made for two assemblies
- In one case, the measured ^{14}C inventory was 25% greater than the revised calculated inventory. In the other case, it was 27% lower
- To obtain a more precise average measured value, a large number of fuel assemblies would have to be analyzed, and about 2/3 of the fuel destined for the repository has not yet been manufactured
- The calculated average inventory is the best value we have at present

Estimate of Repository ^{14}C Inventory

Assumptions

1. Total repository capacity: 70,000 metric tons, of which 7,000 is defense waste and 63,000 is spent fuel
2. Only spent fuel has ^{14}C
3. Average burnups: 29,500 MWd/MTU for BWR
   37,500 MWd/MTU for PWR
4. Percentage on tonnage basis: 35.7% BWR
   64.3% PWR

Result: 1.12 curies per metric ton uranium
or 71 kilocuries total.
Higher fuel burnups than assumed above would result in a proportionally higher ^{14}C inventory

Physical Distribution and Chemical Forms of ^{14}C in Spent Fuel

- For PWR spent fuel with 33,000 MWd/MTU burnup

Component	Inventory (curies/MTU)
UO_2	0.60
Zircaloy	0.18
Fuel assembly hardware	0.22
Total	1.00

- There is a negligible amount in the fuel-rod gas
- About 2% of the overall ^{14}C inventory is readily available on the outside surfaces of the zircaloy cladding (Smith and Baldwin, PNL)
- About 1/2% is readily accessible to the fuel-cladding gap (Wilson, PNL)

Physical Distribution and Chemical Forms of ^{14}C in Spent Fuel

(Continued)

- The chemical forms are not certain, but ^{14}C is believed to exist partially as elemental carbon in the UO_2 and the rest as carbide or oxycarbide
- In the metals, it probably exists as dissolved carbon, but possibly as carbides
- The form in the zircaloy oxide layer is not known

Observations: Release of ^{14}C from Spent-Fuel Cladding

- From measurements performed at LLNL in 1983, it was discovered that ^{14}C is released rapidly from the outside surfaces of intact spent fuel when heated in air and appears as $^{14}CO_2$ in the air (Van Konynenburg et al. 1985)
- Subsequent work by Smith and Baldwin at PNL has shown that as much as 2% of the spent fuel ^{14}C inventory is released from zircaloy cladding in 8 hours at 350°C in air
- The time dependence of the release is consistent with diffusion from a layer of finite thickness, which suggests ^{14}C release from the oxide layer on the zircaloy
- The temperature dependence is Arrhenius, with an activation energy of about 18 kcal/mole

Observations: Release of ^{14}C from Spent-Fuel Cladding

(Continued)

- The release at 200°C is about a factor of 25 lower than at 350°C; at 100°C it is about a factor of 100 lower in 8 hours
- In commercial-grade argon, the release is about a factor of 10 lower than in air. German work indicates that the release is halted by removing essentially all O_2 and H_2O from the argon (Kopp and Münzel, 1990)
- Effects of ionizing radiation on oxidation of ^{14}C have not yet been quantified, but there are indications that carbon is more rapidly oxidized in irradiated air than in non-irradiated air at temperatures as low as 90°C (Reed, ANL)

Observations: Release of ^{14}C from Spent Fuel UO_2

- 50% of ^{14}C inventory in irradiated UO_2 was released within 4 hours of roasting in air at 480°C (Stacy and Goode, 1978)

Observations: Release of $^{14}CO_2$ from Waste Packages

- No release of $^{14}CO_2$ can occur from waste packages, unless the waste containers are breached
- No release of ^{14}C from UO_2 or from the fuel-cladding gaps can occur, unless the cladding is breached

Observations: Release of $^{14}CO_2$ from Waste Packages (Continued)

Examples for a Site Characterization Plan-Conceptual Design (SCP-CD) waste package

- Breach of 1 container in 20,000 in a single year during the containment period, coupled with a 2% loss from this container, would exceed the DOE interpretation of "substantially complete containment"

- Breach of 1 container in 2,000 in a single year during the controlled release period, coupled with a 2% loss from these containers, would violate the NRC 10 CFR 60 limit of 1 part in 10^5 per year

- Groups from LBL and BNL have theoretically analyzed the release of $^{14}CO_2$ through perforations of various sizes in waste containers

Consequences of Total ^{14}C Release

- Amount released would be less than 1/3000 of global ^{14}C inventory
- Three years worth of natural cosmic ray ^{14}C production would exceed the entire repository inventory
- Instantaneous total release, followed by atmospheric mixing, would raise the average atmospheric concentration less than 2%
- Total release over a time longer than a few hundred years would raise the atmospheric concentration less than 0.1% (for comparison, atmospheric nuclear weapons tests raised it by 84% in the northern hemisphere)
- Modeling indicates that the ^{14}C specific activity in the atmosphere will not significantly increase out to the year 2050 from world-wide nuclear energy production, when dilution with ^{12}C from fossil fuel burning is included (McCartney et al. 1988)

Consequences of Total ^{14}C Release

(Continued)

- Average dose to individuals from release of the entire inventory would probably be less than 1 microrem per year, as compared with the average background (including radon) of about 300 millirem per year
- Maximum dose to individuals depends on where food is grown. For reasonable assumptions, it comes out less than 0.1 millirem per year (Dager, 1987)
- Average lifetime fatal cancer risk to individuals would be less than 10^{-8}
- Global population dose would produce an average of one cancer death worldwide per year over 10,000 years. This assumes a population of 10 billion and a linear dose-response, no-threshold relationship down to background dose levels

Information Needs for Improving Our Ability to Model the ^{14}C Source Term

1. Better estimates of time-to-failure for containers and zircaloy cladding
2. Better estimates of number and size distributions of container and cladding perforations
3. More data on release of ^{14}C from zircaloy in air at elevated temperatures
4. Chemical form (or forms) of ^{14}C in spent UO_2
5. Release rate of ^{14}C from UO_2 in air at elevated temperatures
6. Effects of ionizing radiation on release of ^{14}C from zircaloy and UO_2
7. Better estimates of long-term oxidation rates of stainless steel, zircaloy, and nickel alloys
8. Better understanding of behavior of impurity carbon during oxidation of UO_2 and metals

Summary

- The repository ^{14}C inventory is fairly well-established
- There are many uncertainties in proceeding from the inventory to the source term
- The consequences of total release of ^{14}C are very small from the standpoint of an individual; whereas, a sizeable number of health effects can be calculated for global population over a long time period
- Source term uncertainties need to be further reduced. However, the degree of reduction will depend upon the provisions of the new high-level waste regulations. The feasibility of significantly reducing these uncertainties should also be taken into account