UNITED STATES
NUCLEAR REGULATORY COMMISSION
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MEMORANDUM FOR: B.J. Youngblood, Director
Division of High-Level Waste Management

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Subject: NUCLEAR WASTE TECHNOLOGY REVIEW BOARD FULL MEETING ON SOURCE TERM IN PERFORMANCE ASSESSMENT

INTRODUCTION

We attended the NWTRB meeting on October 14-15, 1992 in Las Vegas, Nevada. John Walton and Prasad Nair attended from the CNWRA. Phil Justus and Paul Prestholt attended from the Field Office. The purpose of the meeting was to discuss the concepts, models and supporting data concerning the radionuclide source term for the proposed Yucca Mountain repository. Source term in this context is defined as the release of radionuclides from the engineered barrier to the geosphere. Following the meeting, Colten-Bradley, Ahn, Walton and Justus visited the Yucca Mountain site and on-site laboratories.

Preliminary comments from the NWTRB indicated that they do not believe that there is the proper emphasis on use of total systems performance assessment to direct research needs for the Yucca Mountain repository. DOE is using study plans created 5 years ago rather than current performance assessments. They urged DOE and the other participants to use the results in this way, and to take advantage of participation in collegial meetings.

TECHNICAL PRESENTATIONS

David Stahl of the M&O began the DOE presentations with an overview of source term concepts and definitions. In this presentation, he presented an alternative design for waste packages and the EBS (attached). He suggested that the SCP design of a vertical canister with an air gap is no longer

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preferred. The new design is for a horizontal canister with multiple barriers consisting of steel liners, crushed tuff, stainless steel shot, and iron oxide, to promote water drainage and sorption of released radionuclides. Design temperatures for the waste containers may be higher or lower than those specified in the current SCP design. At higher temperature, computations show convincingly that the containers might remain dry for an extended period, but there are more technical uncertainties about the materials. Conversely, material properties are better understood at low temperatures, but there would be more uncertainty about the rewetting and corrosion of the waste containers. Currently, DOE prefers the higher temperature design. Mathematical models are being developed to predict the behavior of each of the barriers. He next discussed the inventory of spent fuel and glass to be stored in the repository. He stressed that the repository will contain a considerable quantity of spent fuel which has not yet been fabricated, so it is difficult to estimate the properties of this material. It is likely that reactor fuel produced in the future will undergo much higher burnup --up to 60 Gwd/MTU-- than most of the spent fuel already in storage. Laboratory tests on the dissolution of LWR spent fuel and the release of radionuclides have been at burnups of about 30 Gwd/MTU. There needs to be tests performed on higher burnup wastes.

Stahl identified the most pressing information needs for the source term as ^{14}C release mechanisms, spent-fuel oxidation kinetics, release of ^{99}Tc and other soluble species, impact of colloids, and cladding/hardware releases. There was some discussion following Stahl's presentation concerning the importance of ^{14}C releases under the conditions of revised EPA and NRC rules resulting from the changes to the Energy Act.

DOE plans to characterize new nuclear materials and higher-burnup spent fuel. Additional work scheduled (subject to the availability of funds) will include improving the geochemical data base for geochemical simulations, especially those dealing with the source term. The research priorities have not been fully coordinated with repository performance assessment modeling however. Dale Wilder of LLNL commented that there will be some work determining how geochemical simulation codes can be used in source term modeling, and where kinetic effects are likely to be important. Work is continuing at PNL on spent fuel dissolution, and DOE may reinstate glass testing. There may also be testing for releases of radionuclides (particularly ^{14}C) from zirconium cladding and other hardware components of the fuel.

Richard Van Konynenburg of LLNL made a presentation of ^{14}C releases from the engineered barrier. ^{14}C is made by neutron activation in the core, primarily by reaction with nitrogen, but also from activation of ^{17}O and ^{13}C. Most ^{14}C produced in LWRs is released to the atmosphere during operation under currently acceptable release limits. The inventory of ^{14}C in the spent fuel is divided between several compartments, including the crud and outer cladding oxide, grain boundaries, cladding gap and UO_2. The current best estimate of ^{14}C in LWR fuels with average burnup of 30 Gwd/MT is about 1 curie/MTHM. Mic Apted commented that the ^{14}C inventory in the fuel would increase with increasing burnup. About 2.5% of the inventory can be expected to be released quickly upon canister failure, but much smaller quantities would be released subsequently from the fuel. The largest inventory is in the UO_2 itself, but Van Konynenburg presented some data indicating that a substantial fraction (up to half) of this inventory would not be released from the fuel because it is not in a form readily oxidized. He added that the consequences of total release of ^{14}C are very small from the standpoint of dose to an individual, whereas a sizeable dose could be calculated for exposure to the global population.

Ray Stout from LLNL presented an overview of spent fuel modeling concepts and material characterization. Fuel and cladding have been tested over very limited ranges of environmental conditions and for short times only. They need to find ways of testing materials to include all of the likely conditions to be encountered in the repository. Nevertheless, the "Preliminary Waste Form Characterization Report" is in preparation to integrate data collected so far. Someone asked how reactor fuel more highly enriched than today's fuel (as a result of purchasing Russian nuclear material) would be treated in the source term. DOE has not yet taken this possibility into account, nor are highly enriched fuels such as naval reactor fuel being considered in performance of the repository.

Robert Einziger of PNL presented results of tests on the oxidation of spent fuel, primarily from work on dry cask storage in air. These results were similar to those presented last February in Pasco WA, and have subsequently been used in the NRC Phase 2 Iterative Performance Assessment to define the gaseous ^{14}C release model. Much of the data are from experiments started up to a decade ago, and on fuels that are of average to low burnup. There are tentative plans to restart the experiments with additional spent fuel. There also might be some direct measurements of ^{14}C from the oxidizing spent fuel that would be very useful for predicting the release rate of this gas for the performance assessments. This was not possible in the previous experiments because radiation from ^{14}C is difficult to measure.

Walter Gray, also of PNL, presented information about the dissolution testing of spent fuel in a flow-through apparatus which allowed relatively large quantities of water to flow over grains of spent fuel without allowing secondary phases to precipitate. The tests were motivated by need for thermodynamic data involving low-solubility radionuclides and the inventories of soluble radionuclides that accumulate at gap and grain-boundaries. The experiments are not typical of the way that spent fuel and water would interact in the repository environment, but could help to establish upper limits of fuel dissolution. Future goals include a thermodynamic data base for components of spent fuel, and to determine the nature and importance of actinide-bearing colloids.

John Bates of Argonne presented an evaluation of colloid formation from glass waste forms. In humid air there can be reactions between a thin film of water and the waste form, leading to an aged glass. Layers of glass can detach from the bulk glass waste form. Colloids can be formed from intermittent wetting of glass by spallation. These colloids may be transported more readily than dissolved radionuclides. William Bourcier of LLNL presented results of geochemical models of the reaction of the glass waste form with water. Donald Langmuir of the NWTRB questioned whether the use of J-13 water as the representative fluid was correct in either the model or experiments. Bates commented that the experimental data used water that was in direct contact with the tuff at 90°C, and therefore was not simply J-13 water.

Following the presentation, Codell discussed a recent paper of which Bates was an author (Wronkiewicz et al, 1992) in which unirradiated UO_2 was subjected to dripping water at 90°C. Bates contends that the static and flow-through tests currently being performed at PNL on spent fuel are fundamentally different from the dripping tests. He urges DOE to perform these dripping tests on actual spent fuel, but it is not clear that this will occur.

Cynthia Palmer of LLNL presented information on the thermodynamic data base for radionuclides associated with the source term. They are placing highest priority on determining thermodynamic data on americium, plutonium, neptunium, uranium, technetium, zirconium and nickel. Much of the results presented was from computer models rather than wet experiments. Don Langmuir stated that the experiments would have been more meaningful if water in equilibrium at high temperature had been used instead of J-13 water. Warner North of the NWTRB brought up the issue of mobilization of radionuclides by organic matter, both natural and man-made. Ardith Simmons of the DOE responded that experimental work on complexation with organics and the movement of radionuclides in colloids will begin by the Spring of 1993.

Thomas Wolery of LLNL presented a description of the geochemical code EQ3/6. This is the main geochemical code being used in the DOE Yucca Mountain investigations. The code is available for Sun workstations and can be ported to PC's. It is the only Yucca

Mountain Project SES undergoing quality assurance procedures. We hope to acquire this code at NRC once the Sun work stations are delivered.

The last formal presentation of the day was by Diane Harrison of the Yucca Mountain Site Characterization Project, DOE, on plans for future work. Initiatives include a new experimental project directly related to characterizing the near field environment, a large($27m^3$) block lab heater test, and verification of the V-TOUGH two-phase flow code, including modification to include coupled matrix/fracture flow and heated-drift stability.

October 15 was devoted mostly to presentations on source term models for use in total systems performance assessments. The first speaker was William O'Connell from LLNL. The LLNL source term model includes most of the concepts that have been developed over the last decade. The model has a relatively simple representation of radionuclide releases that can be run quickly for total systems performance assessment models, but lacks much of the mechanistic detail of some models. They contend that the present source term model is good enough for early system performance assessments. More sophisticated models and data will be included in future performance assessments.

Mike Wilson and Rally Barnard of Sandia presented the applications of the LLNL source term model to the DOE Total Systems Performance Assessment (TSPA) for 1991. The TSPA takes into account two modes of flow relative to the source term; either the "composite porosity" or "weeps" models. The former model considers that the tuff in contact with the canister is wet, and all canisters are therefore contacted. The latter model assumes that only some of the containers come into contact with the water by means of dripping fractures or "weeps", and only those canisters fail and release their radionuclides. The weeps model accounts for greater release and transport of aqueous radionuclides than the composite porosity model, but less gaseous release. Future phases of their performance assessment will include new container and emplacement concepts and more mechanistic models for behavior of the waste.

David Engel presented the PNL source term model embodied in the code SUMO. This code like the other DOE source term codes deals mostly with simple conceptualizations. They devote more time to thermal modeling and how it affects resaturation of the repository, but there are few instances of mechanistic modeling of the release rate mechanisms. They have separate models for glass and spent fuel waste forms. They plan to couple geochemical codes into their analysis to determine the aqueous environment for canister corrosion, but this has not yet been implemented.

Robert Shaw presented the EPRI source term model for their total system performance assessment. The EPRI performance assessment differs from the DOE and NRC performance assessments in the way they deal with scenarios and parameter uncertainties. The EPRI model uses individual experts to develop nodes on a logic tree, and determine the degree of belief in the branches at each node. This differs from the more traditional methods of scenario identification coupled with parameter values sampled with Monte Carlo methods. The EPRI source term model itself is largely non-mechanistic, using experts to assign failure rate and other characteristics for the barriers. This procedure was even extended to the temperature of the canisters, even though we believe that temperature is not really a probabilistic variable.

The EPRI model considered dry, wet drip and moist conditions for release from the waste, and either fuel alteration rate or solubility limited release. While they recognize temperature as being potentially important to release, they did not include this dependence in their models because the state of knowledge of parameters such as solubility is so poor.

Most of the DOE and EPRI presenters acknowledged the potentially important impact of changes in the design of the repository, state of the waste form and regulations on performance assessment needs. DOE seems to be leaning toward an emplacement strategy which uses containers placed horizontally in drifts rather than in vertical boreholes with liners and air gaps. Such a change in design would have a profound impact on the source term model. Bob Shaw mentioned the use of a universal casks which is sealed at the power plant and used for both shipping and burial. This also would be quite different from the SCP design.

Changes to the high level waste regulations because of the new Energy Act legislation may obviate the need for detailed analysis of the ^{14}C releases. Standards based on individual doses rather than cumulative release might require more complicated and detailed source term and transport models.

Codell presented the status of the source term model being used for Phase 2 of NRC's iterative performance assessment. A copy of the overheads is attached.

Mic Apted, a consultant to the NWTRB, presented the status of a survey of models for high-level waste repositories, and plans for an international workshop on source term models to be held in May 1993. He made a point of the need for site data on diffusion in unsaturated media under repository conditions, making the claim that it might be possible to gain a great deal of credit for the apparently low diffusion through dry or nearly dry materials.

Nava Garisto, another consultant to the NWTRB, gave a presentation on the Canadian perspective on source term modeling. The Canadian repository is quite different in environment; e.g., saturated Canadian Shield granite vs. unsaturated tuff, reducing vs. oxidizing conditions, titanium or copper containers vs. stainless steel, and CANDU spent fuel vs. light water reactor spent fuel and glass. Nevertheless, the basic approach to the source term is similar in many respects to those of the U.S. programs.

R. Spengler (USGS) discussed the recent mapping of the Ghost Dance Fault by the U.S. Geological Survey. The fault is currently being mapped at a scale of 1:50. The fault zone is now considered to be as wide as 700 feet. Further mapping of the fault and related features is planned for FY 1993.

Carl Gertz presented the YMP budget for 1993. There was emphasis of the priority to get underground during the 1993 fiscal year. Construction of the ESF is expected to start in November. The TBM has not yet been delivered. The NWTRB expressed concerns about the low drilling rates for the LM300 (average of 7 ft/day).

FIELD TRIP TO THE REPOSITORY SITE

On October 16, John Walton, Tae Ahn, and Virginia Colten-Bradley visited Yucca Mountain with NRC field representative Phil Justis. Stops included the YMPO core-handling facility, the USGS WRD labs, the site of the North Portal and Midway Valley Trench, the LM300 drilling rig, the escarpment along the Ghost Dance Fault, Trench 14, Busted Butte, J-13 well, and top of Yucca Mountain. Phil gave a well-prepared and very informative introductory site visit, with handouts, maps, etc., and lunch in the shade.

CONCLUSIONS AND RECOMMENDATIONS

The NRC and CNWRA staff in attendance generally felt that the meeting was worthwhile. It gave us an opportunity to see the latest results coming from DOE-funded laboratories and the state of development of models for the release of radionuclides from the waste form. The models presented for use in the DOE total systems performance assessments do not appear to be more highly developed than NRC's models, and in the area of ^{14}C releases and container failure, less developed. One of the keys to better models is better and more-extensive experimental results. Only DOE is currently equipped to perform experiments on irradiated fuels, but funding of these experiments is severely curtailed. The NWTRB urged DOE to let the results of total systems performance assessments prioritize and direct how the limited experimental budgets are spent, rather than relying on test plans developed years ago. The position of DOE is in apparent agreement with the NWTRB recommendation in part, with an emphasis on the use of performance assessment and expert judgement for setting priorities in their "Mission 2001" plan.

We have all handouts from the meeting, and would be happy to brief you further.

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Enclosure:
1. Figure of an alternative concept for engineered barrier
2. NRC presentation overheads on source term

DISTRIBUTION:

Central File	BJYoungblood, HLWM	JJLinehan, HLWM	HLHP r/f
MVFederline, HLHP	RLBallard, HLEN	JHolonich, HLPD	NMSS r/f
RCodell, HLHP			

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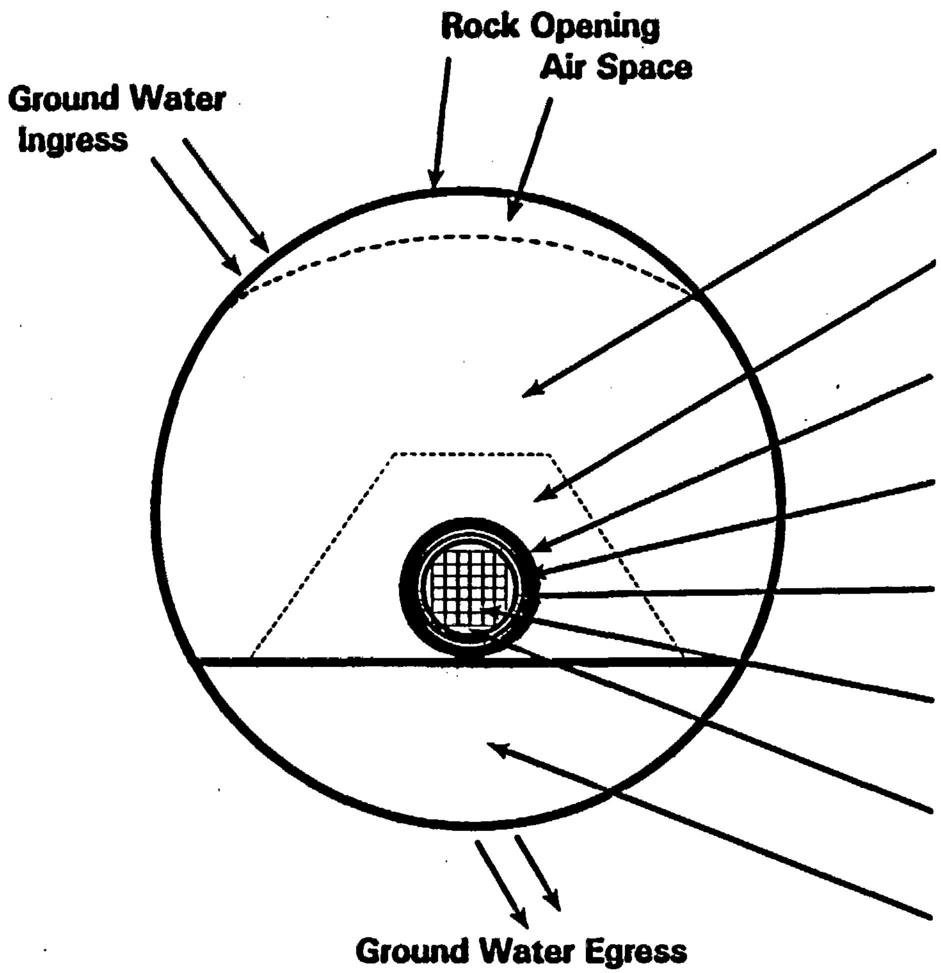
REFERENCE

Wronkiewicz, D.J., J.K. Bates, T.J. Gerding, E. Veleckis, and B.S. Tani, "Uranium release and secondary phase formation during unsaturated testing of UO_2 at 90 °C", J. Nuclear Materials, 190 (1992), pp 107-127

Waste Package/EBS Performance Assessment of <u>One</u> Design Concept



Components	Possible Materials
Engineered Backfill	Crushed Tuff
Engineered Packing	Coarse Tuff
Outer Containment Barrier	Weathering Steel
Separator	Stainless Steel Shot
Inner Containment Barrier	Alloy 825
Spent Fuel/Basket Assembly	Borated Material
Buffer/Filler Material	Iron Oxide Powder
Engineered Invert	Coarse Tuff

Source Term Model for USNRC Iterative Performance Assessment, Phase 2

Richard Codell, USNRC
Tae Ahn, USNRC
John Walton, CNWRA

Nuclear Waste Technical Review Board
Las Vegas, Nevada
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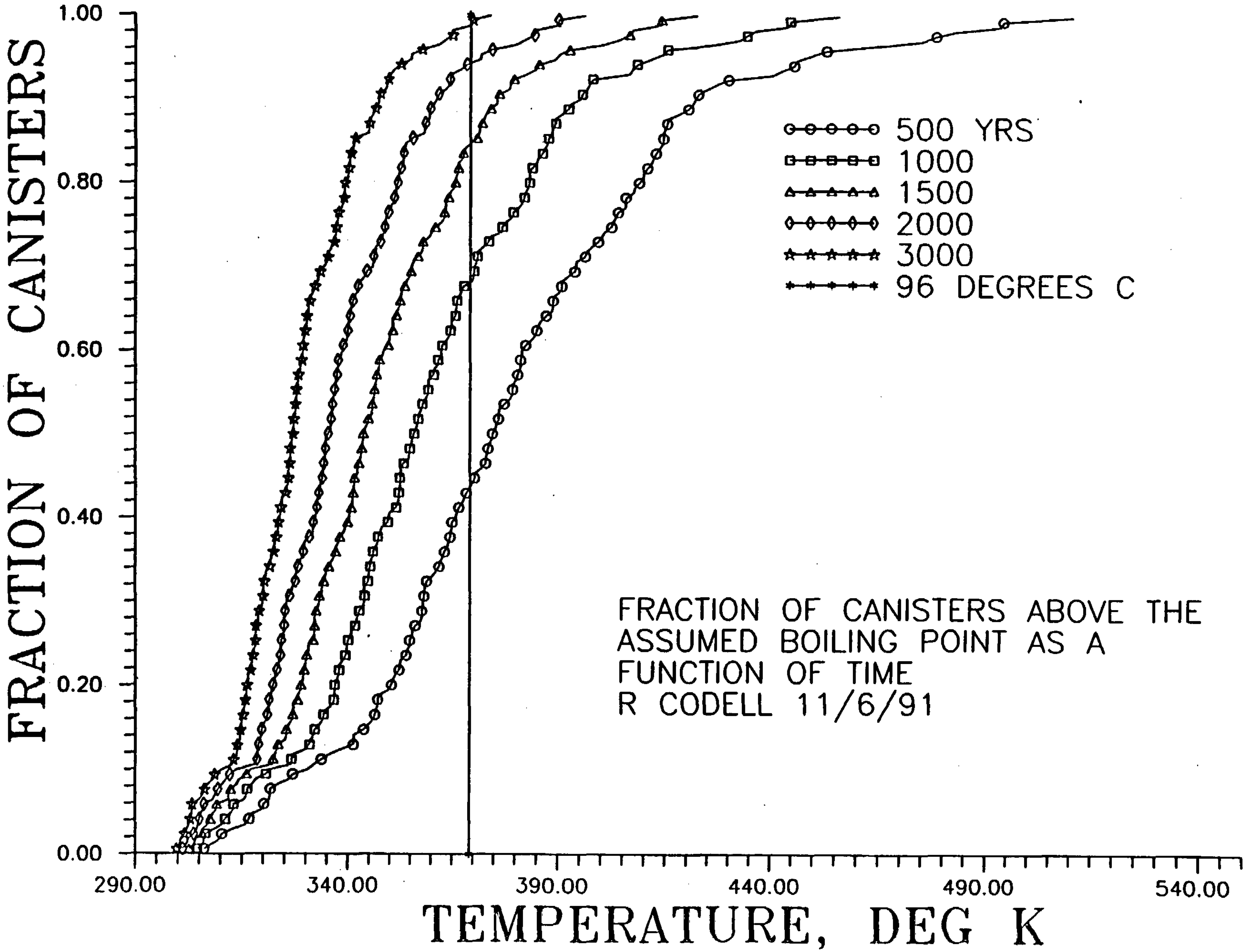
Canister Temperature Model

- Semi-analytical, conduction only
- Uniform heat transfer properties
- Heat load can vary in time and space
- Mainly used to determine time to canister failure



FRACTION OF CANISTERS
1.00
0.80
0.60
0.40
0.20
0.00
290.00
340.00
390.00
440.00
490.00
540.00
TEMPERATURE, DEG K
500 YRS
1000
1500
2000
3000
96 DEGREES C
FRACTION OF CANISTERS ABOVE THE
ASSUMED BOILING POINT AS A
FUNCTION OF TIME
R CODELL 11/6/91

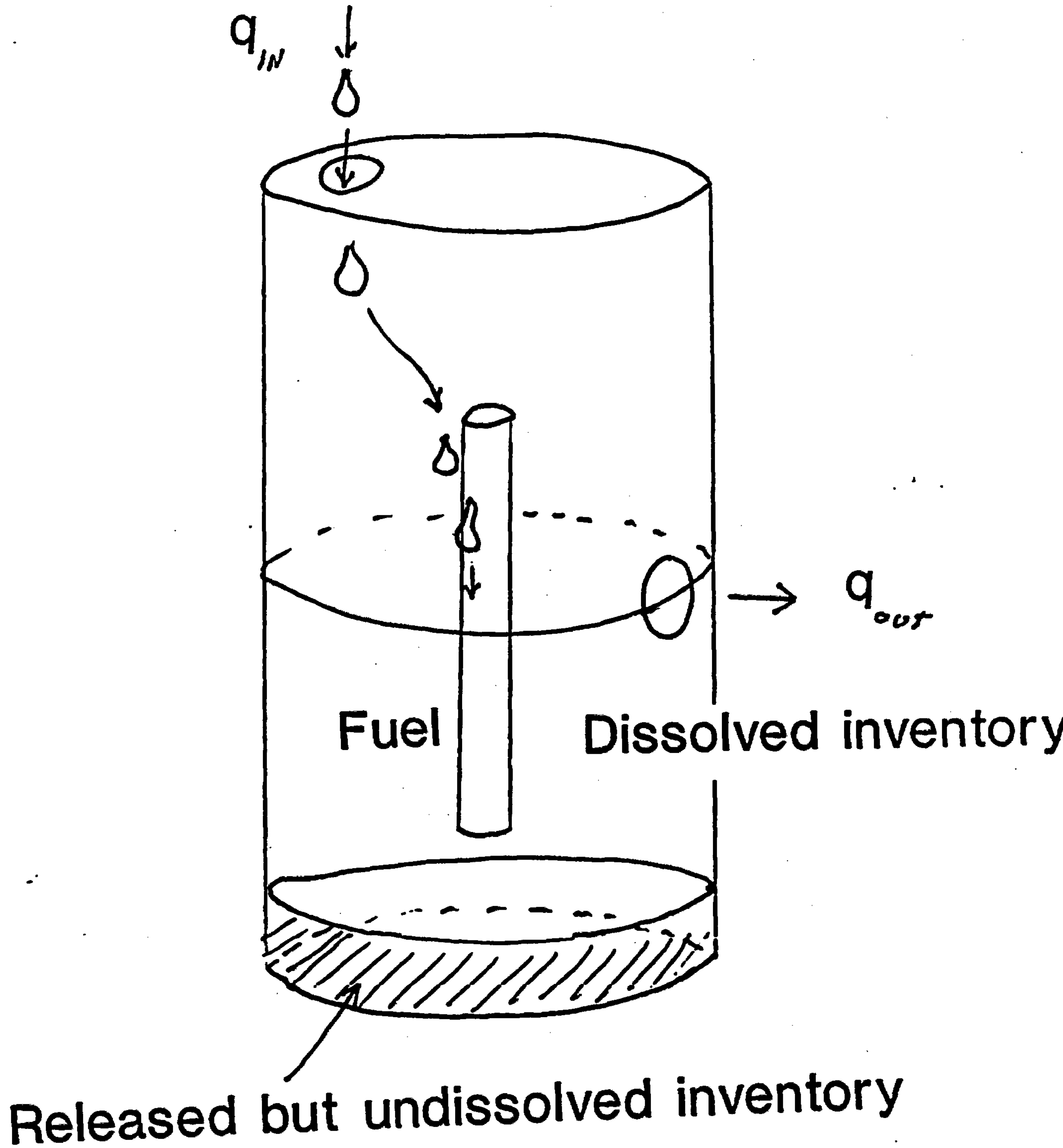
Canister Corrosion Model

- General corrosion
- Crevice corrosion
- Pitting

Other Canister Failure Modes

- Buckling
  - SCP design, 304L Stainless
  - Long cylinder
  - Thickness decreases by corrosion
  - No air gap
  - No stiffening
- Initial defectives
- Disruptive scenarios
  - Seismic failure
  - Volcanism
  - Drilling

Dissolved Radionuclide Release Model

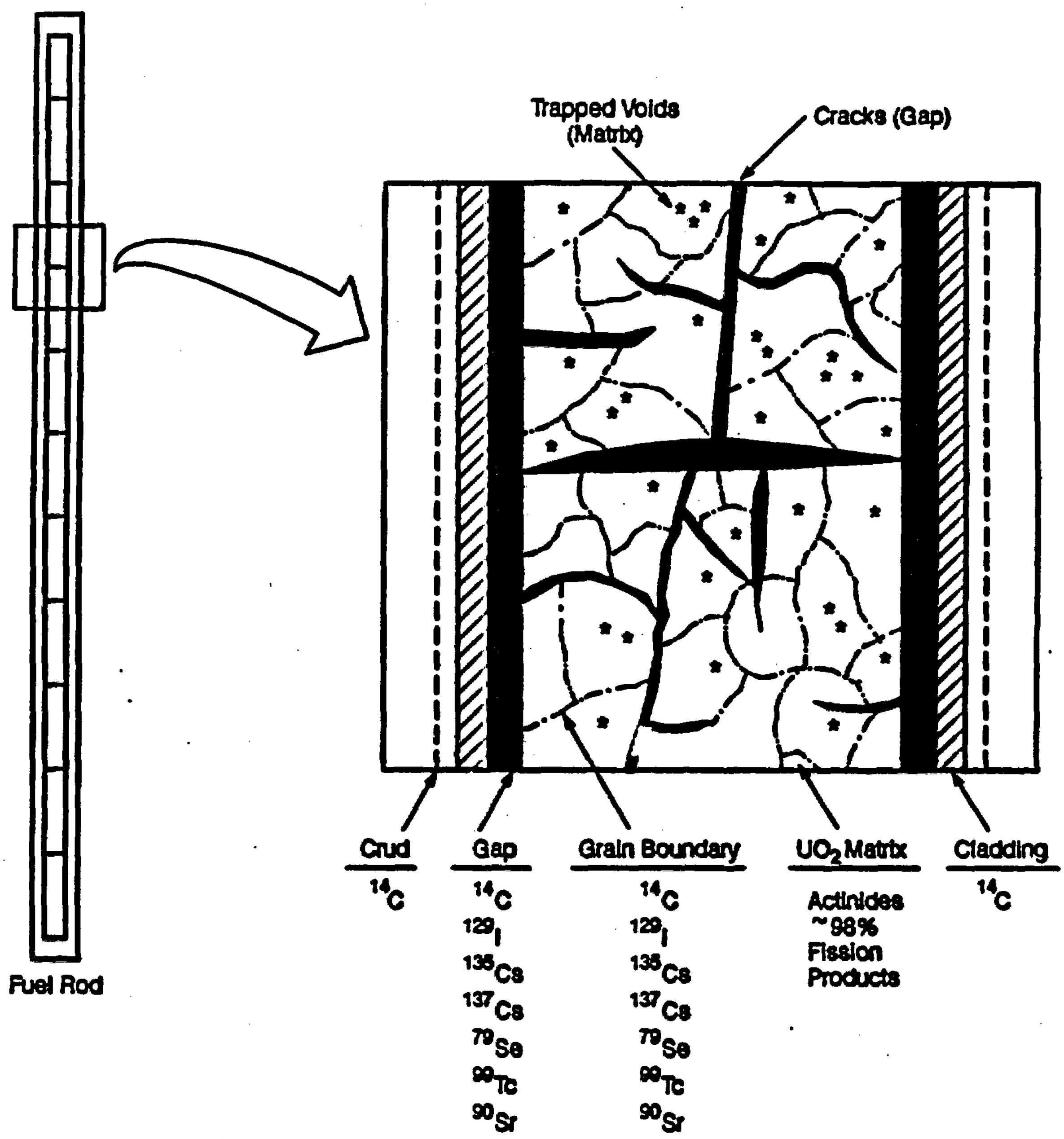


Plutonium Releases

- Pu and Am dominate dose potential, but
- Very insoluble - Largely ignored in other performance assessments
- Kinetic effects may play an important part in releases of Pu and other actinides
- Potential concentrations of Pu taken from data by Nitsche et al, and Wilson et al, E-5 to E-9 M
- Speciation calculations show range reasonable at 25°C (not at 85°C)

EVIDENCES FOR KINETIC EFFECTS

- MULTI-PHASE FORMATION
- PARAGENESIS OF SECONDARY PHASES
- UNSTABLE SECONDARY PHASES
- NONPROTECTIVE SECONDARY PHASES
- ENVIRONMENTAL CHANGES



Location of Radionuclides in Spent Fuel and Potential Releases of C–14
(Apted, el. al., 1989)

Model for Release of C-14

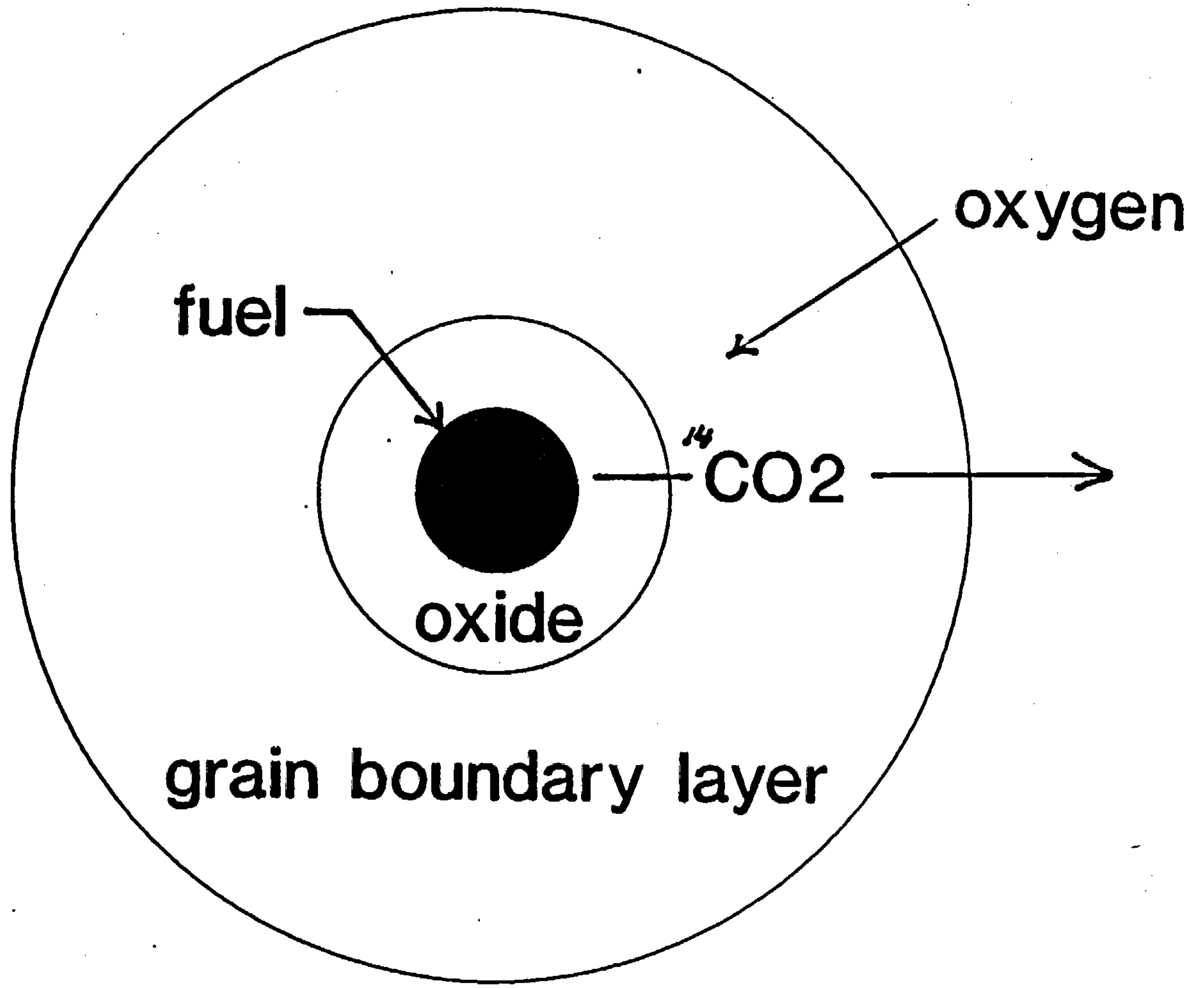
- C-14 is in a reduced state initially in fuel
- C-14 oxidizes to carbon dioxide as fuel oxidation front passes
- C-14 dioxide diffuses out through same two layers as oxygen diffuses in
- C-14 released quickly from grain boundaries, cladding/fuel gap, and initial zirc. oxide
- Minor releases from oxidation of cladding and other metals

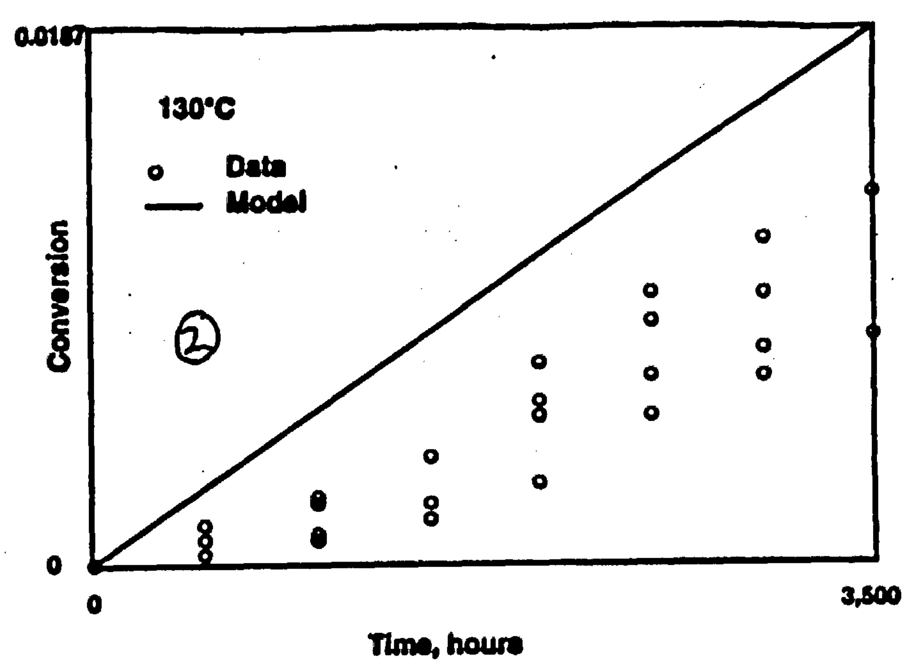
Fuel Oxidation Model

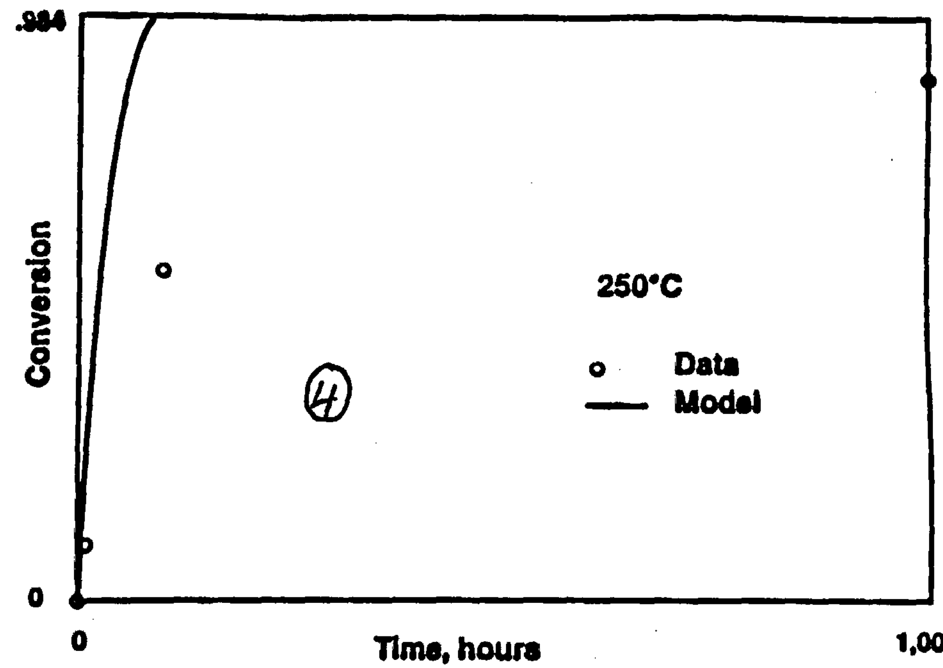
Assumptions:

- No oxidation until canister fails
- No protection of fuel by cladding
- Oxygen diffuses through two layers:
  - outer layer representing grain boundaries
  - inner layer representing oxidized fuel
- Oxide is U3O7 stochiometrically
- Oxygen concentration zero at inner boundary
- Oxygen profiles in layers are at steady state

C-14 Gaseous Release Model




0.0187
130°C
Data
Model
Conversion
0
0
3,500
Time, hours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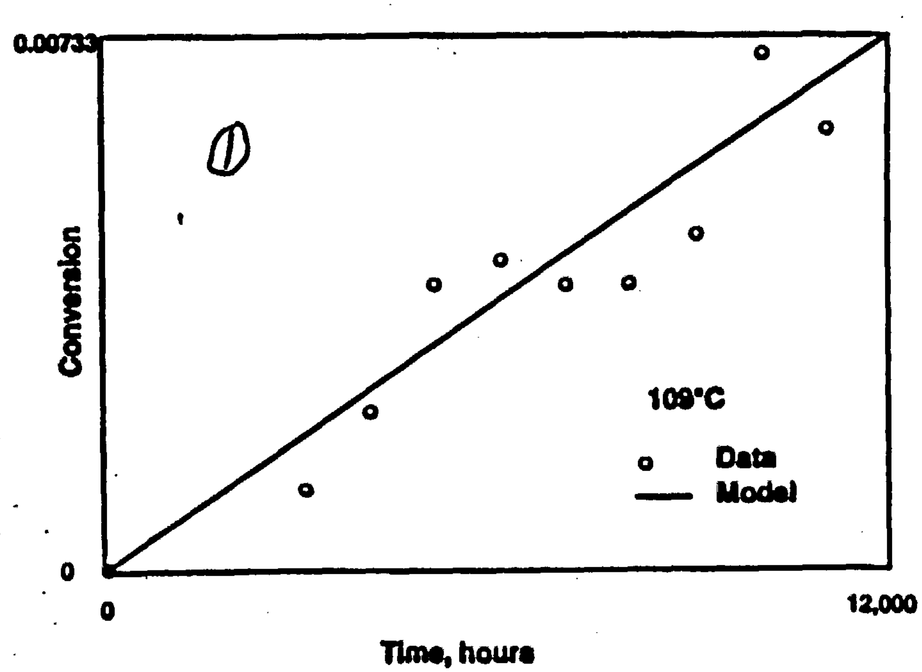
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Conversion
109°C
Data
Model
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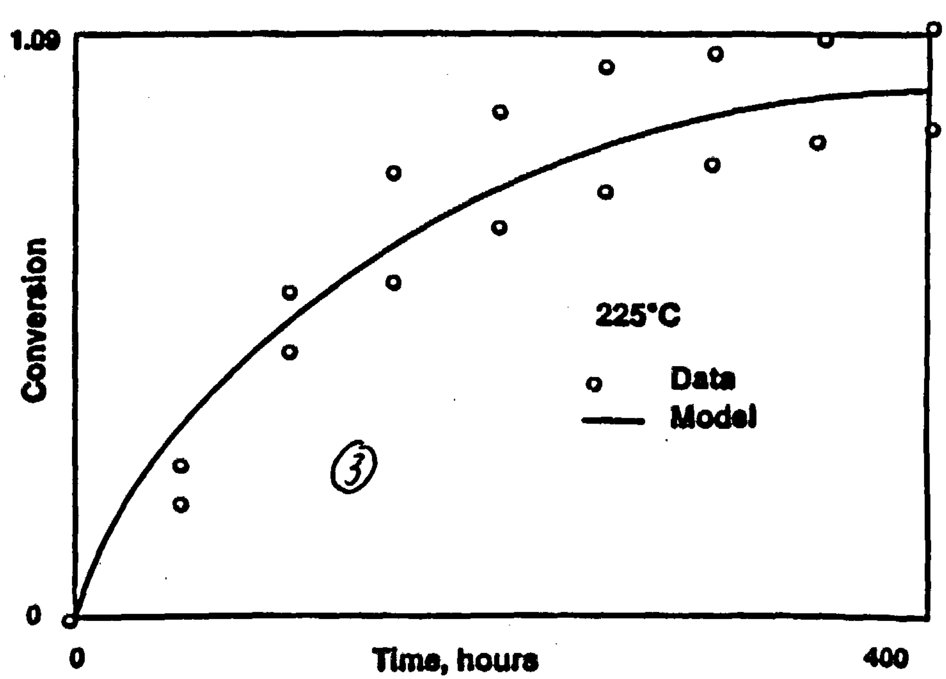
0
12,000
Time, hours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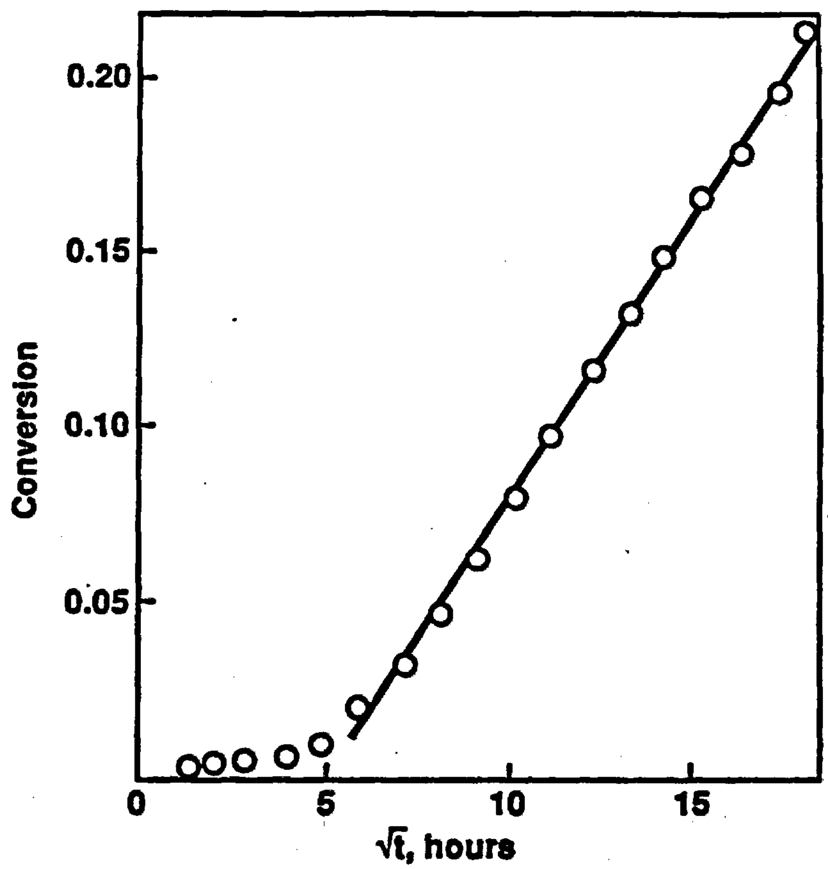
Time, hours

Transient Oxidation Effects



Not included in fuel oxidation model

Disruptive Releases

- Intrusive Volcanism
  -Dike 1000 - 4000 m long, 1-10 m wide
- Extrusive Volcanism
  - Cindercone, 25 - 100 m radius
- Drilling intercepts repository
  - Brings up contents of waste package
  - Brings up contaminated rock

VAPOR PRESSURES

SPECIES	**VAPOR PRESSURE (ATMOSPHERES)**	
	100°C	**200°C**
CO_2	> 2,000	> 12,000
I_2	6×10^{-2}	3.7
SeO_2	9.1×10^{-4}	5.4×10^{-2}
Tc_2O_7	1.2×10^{-4}	3.7×10^{-2}

(FROM LANGE'S HANDBOOK OF CHEMISTRY, 13TH EDITION, 1985)

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Conclusions

NRC SOTEC model includes:

- Waste package failure
- Release of liquid (dissolved) radionuclides
  - Colloids treated by empirical data
- Gaseous releases of C-14

Other analyses and codes for:

- Other gaseous nuclides
- volcanic intrusions
- drilling