UNITED STATES
NUCLEAR REGULATORY COMMISSION
WASHINGTON, D. C. 20555

SEP 18 1985

MEMORANDUM FOR: John G. Davis, Director
Office of Nuclear Material Safety
and Safeguards

FROM: Robert B. Minogue, Director
Office of Nuclear Regulatory Research

SUBJECT: RESEARCH INFORMATION LETTER # 143 - GASTROINTESTINAL ABSORPTION OF PLUTONIUM IN MICE, RATS, AND DOGS

Introduction:

This memorandum transmits the results of experimental studies of the gastrointestinal absorption of plutonium in mice, rats, and dogs. The results have direct application to calculation of doses from plutonium ingestion. The research project was initiated in response to a request from the former Office of Standards Development, RR-SD-79-6, to determine the fraction of radionuclide transferred from gut to blood. This fraction, f_1, is used in calculating doses to internal organs resulting from ingestion of plutonium. Several investigators, working independently, have shown that the previously accepted value (ICRP-2) for f_1 (1×10^{-5}) was incorrect because the Pu(IV) used in the earlier experiments hydrolyzed, a phenomenon which does not always occur, and which reduces solubility. Consequently, the calculated doses were too low. In recognition of this fact, the International Commission on Radiological Protection in 1979 raised its value to 1×10^{-4} (The National Radiological Protection Board in England later adopted a value of 5×10^{-4}.). However, the subject experimental results suggest that 1×10^{-4} is still too low by a factor of ten. The work was performed by Drs. Maryka Bhattacharyya and Robert Larsen of the Division of Biological and Medical Research at the Argonne National Laboratory. The report describing these studies, entitled "Gastrointestinal Absorption of Plutonium in Mice, Rats, and Dogs: Application to Establishing

143

Values of f_1 for Soluble Plutonium," NUREG/CR-4208, has been transmitted to your staff.

Methodology:

Mice and rats were exposed to solutions of plutonium either ad libitum via drinking water or by gavage (direct administration into the stomach). Plutonium solutions were administered to dogs via gelatin capsules. Plutonium concentrations were 1×10^{-10} M, the molar concentration of Pu-239 at the MPC, the maximum permissible concentration in drinking water (5 pCi/ml). The administration medium was 0.01 M sodium bicarbonate, a medium similar to Lake Michigan water. The oxidation state was, except where noted, Pu(VI), the oxidation state present in chlorinated drinking water. At sacrifice of mice and rats, between 2 and 6 days after administration, pelts were removed, bodies were eviscerated, and plutonium was assayed in lungs, livers, and skinned eviscerated carcasses. Dogs were sacrificed 5 weeks after administration and their pelts and gastrointestinal tracts were removed; livers, lungs, and skeletons were removed and assayed. Plutonium concentrations in tissues were determined by gamma spectroscopy for Pu-237 and by alpha spectrometric isotope dilution for Pu-236, Pu-238, and Pu-239.

Results and Discussion

1. Effect of Plutonium Oxidation State

To determine the effect of the oxidation state of plutonium on GI absorption, two groups of adult female mice were fasted for 24 hours and administered Pu(IV) or Pu(VI) by gavage. The fraction of plutonium

retained at 6 days was 2×10^{-3} and was the same for both Pu(IV) and Pu(VI).

2. Effect of Plutonium Concentration

To determine the effect of plutonium concentration on the GI absorption of plutonium, three groups of fed mice were administered Pu-236 at a concentration of 1×10^{-12} M, Pu-238 at 1×10^{-10} M, and Pu-239 at 1×10^{-8} M. The fraction of Pu(VI) retained by fed mice at 21 days was independent of concentration over four orders of magnitude.

3. Effect of Administration Medium and Hydrolysis

To determine the effect of administration medium, tetravalent Pu-237 was administered to three groups of fasted adult female mice as nitrate, citrate, or bicarbonate solutions. The retention of Pu-237(IV) was not significantly affected by the nature of the anionic species of the solutions.

Additional experiments examined the effect of hydrolysis of Pu(IV) characterized by the degree of ultrafilterability (low UF values indicate polymerization). A decrease from 70% UF to 10% UF resulted in a decrease in GI absorption from 2×10^{-3} to 3×10^{-4} in the fasted mouse.

4. Effect of Animal Species

To determine the effect of animal species on the GI absorption of plutonium, a solution of Pu(VI) was administered to fasted mice by gavage, to fasted rats via drinking water, and to fasted dogs via gelatin capsule.

The fraction of plutonium retained at 6 days was 1.5×10^{-3} for mice, 3.2×10^{-3} for rats, and 6.6×10^{-4} for dogs at 5 weeks.

5. Effect of Feeding Regimen and Time of Administration

To determine the effect of food deprivation on the GI absorption of plutonium, a group of adult male mice was administered Pu-236 via drinking water with food available and 2 days later Pu-238 via drinking water following a fasting period. The fraction of plutonium retained 7 to 9 days after GI absorption of Pu(VI) in the fasted mouse (1.9×10^{-3}) was 13-fold higher than in the fed mouse (1.4×10^{-4}). A similar experiment showed that uptake during the active cycle (night) was the same as during the inactive cycle (day).

6. Effect of Age of the Animal

To determine the effect of the age of the animal, Pu-238 solutions were administered to groups of fed rat pups who were 1,2,10,19,29,52, and 100 days old. The fraction of Pu-238 retained by the fed rat at 1 day of age was 7.4×10^{-3}, seventy-fold higher than the value of 1×10^{-4} for the 100 day old adult. At 19 days, just prior to weaning, the fraction retained was 3.1×10^{-3}, approximately the same as the fasted adult rat. By 29 days, 1 week after weaning, the fraction retained had decreased to adult levels, 1×10^{-4}.

Conclusions and Recommendations

The objective of these studies was the determination of f_1, the fraction of radionuclide transferred from gut to blood. Two cases must be considered for application: workers and the general population.

For consideration of exposure to plutonium in the workplace, the most likely situation would be an accident; i.e., a single exposure. However, two groups of individuals are considered: those who eat a normal breakfast and those who skip breakfast. For the fed worker, a value of f_1 of 2×10^{-4} is recommended based on the results for the fed adult mouse. For the fasted worker, a value of 2×10^{-3} is recommended.

In considering the environmental exposure to plutonium in drinking water, two approaches were developed. The first involved calculating the amounts of Pu-239 and the doses accumulated with age for a concentration of 1 Bq/l.

Age-dependent values of f_1, fluid consumption, and body weight were used to produce the curves given in Appendix C of NUREG/CR-4208. A single f_1 was determined by calculating the value that would give the same lifetime dose.

The second approach involved calculating the dose commitment over a 70-year lifetime from each year's uptake of Pu-239 from drinking water containing Pu-239 at 1 Bq/l. Since the first year of human life is the year during which uptake of plutonium from drinking water results in the greatest commitment of lifetime dose (14.4%) due to high GI absorption, low body weight, and long residence time, a single value of f_1 for the first year of life is limiting.

Using the first approach, an $f_1 = 3 \times 10^{-4}$ would protect fed individuals of all ages and $f_1 = 6 \times 10^{-4}$ would protect fasted individuals. The second approach, which limits exposure by applying an ALI (annual limit on intake) to the first year of human life, would produce an $f_1 = 4 \times 10^{-3}$.

We recommend that NMSS consider the above f_1 values in conducting regulatory assessments. With regard to NRC regulations (10 CFR Part 20), we are proposing

to adopt current ICRP intake recommendations (f_1 of 1×10^{-4} for class D plutonium). This question will be reviewed following confirmation of the results of this study through ongoing experiments using a non-human primate.

Future Work

In collaboration with investigators at New York University, four baboons have been administered isotopes of plutonium, uranium and neptunium. Results should be available in FY 1986. At that time we expect to inform ICRP of the results and to initiate any necessary action to change the maximum permissible concentration for plutonium in liquid effluents. For further information, please contact Dr. Judith D. Foulke (427-4563).

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Original signed by:
ROBERT B. MINOGUE

Robert B. Minogue, Director
Office of Nuclear Regulatory Research

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MEMORANDUM FOR: Harold R. Denton, Director
Office of Nuclear Reactor Regulation

FROM: Robert B. Minogue, Director
Office of Nuclear Regulatory Research

SUBJECT: RESEARCH INFORMATION LETTER, NO. 142 "RELIABILITY ANALYSIS OF STIFF VERSUS FLEXIBLE PIPING"

Introduction

Current design criteria for piping evolved under the presumption that higher seismic margins necessarily improve plant reliability. Conservative design against earthquake loads has relied increasingly on rigid supports, snubbers, and other types of seismic restraints to stiffen piping systems. The resultant decrease in flexibility, however, is likely to cause higher normal operating stresses because of the restraint of thermal expansion. Furthermore, because of the large uncertainty inherent in predicting seismic effects (compared to that in predicting thermal effects), seismic loads dominate the design even though seismic loads occur very infrequently. As a result, stiffening a piping system to improve its resistance to seismic loads may actually decrease its overall reliability during normal operation.

In order to quantify piping reliability, pipe failure probabilities were computed by Lawrence Livermore National Laboratory. Pipe failure was assumed to be caused by fatigue crack growth at pipe weld joints. Two types of failure modes were considered initially, i.e., a through wall crack (leak) and a complete pipe severance (break). Pipe support and on-line component failure modes were subsequently introduced. Studies were undertaken to evaluate changes in reliability of pipe supports and on-line components as a function of piping flexibility based on available fragility data. Pipe failure probability was estimated by applying a Monte Carlo method with a stratified-sampling scheme to simulate the life histories of the piping system.

Selected piping systems with related design data were collected from real nuclear power plant designs. Flexible piping designs were created from the existing designs by removing rigid supports and/or snubbers. Piping stresses for various designs were calculated for the reliability assessment.

This assessment recognizes the characteristic difference between regular pipe supports and snubbers. While removal of regular pipe supports changes both seismic stress and thermal expansion stress in a piping

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system, removal of snubbers affects only seismic stress. However, a piping system including snubbers may not exhibit the desired reliability because snubbers are known to have a high failure rate. The possibility of snubber malfunction is incorporated in this assessment.

The malfunction of pipe whip restraints was also treated. The situation where the pipe comes in contact with a restraint device during normal operation due to an imperfect installation was investigated. Actual stresses caused by the malfunction were calculated and an assessment of the safety impact on the piping was undertaken by performing a reliability analysis with and without the malfunction. The intent was to confirm that the malfunction of pipe whip restraints introduces higher thermal stresses and reduces the overall piping reliability.

Results

Based on this research, the change in piping reliability was determined to be largely insensitive to the change in piping flexibility for the cases studied. The piping reliability is either improved or affected very little by the increased piping flexibility as a result of removing rigid pipe supports and/or snubbers. Pipe failure probabilities are generally small; values of the order of 10^{-9} per reactor year are reported for pipe rupture and 10^{-6} per reactor years are reported for pipe leak. It is concluded that flexible piping design is desirable based on reliability considerations, and that flexible piping design also offers many other benefits.

Although it has been demonstrated that piping systems can be made more reliable by adopting flexible piping designs, increase in piping flexibility usually results in greater pipe displacements. Displacement criteria or requirements to confine piping displacements may be needed.

Since pipe supports and on-line components (such as pumps and valves) are important parts of a piping system, the effects of increased piping flexibility on the reliability of supports and components were investigated. Results indicated that the support reliability usually exhibits a moderate decrease as the piping flexibility increases. It is felt that the supports in a flexible piping design need to be either upgraded or subjected to further investigation.

For large components, such as steam generators, pressurizers, and large pumps, the global effect concerning the component support failure due to increased nozzle loads was evaluated. It was found that in general the global effect is rather insignificant. However, the local effect at the vicinity of the nozzle may need to be further investigated. Without such an investigation, it is suggested that removing pipe supports which are close to nozzles should be done with extreme care.

For self- supporting on-line valves, it was discovered that the valve acceleration may or may not increase with piping flexibility. Nevertheless, valves usually have sufficient design margins to accommodate the higher acceleration and are able to maintain functionality. The problem in this case is the increased valve displacement usually associated with a flexible piping design. Specific design consideration may be needed in order to limit the valve displacement.

Malfunctioning pipe whip restraints were found to increase thermal stresses by a factor of 15 in the worst case; this malfunction increases pipe rupture probabilities by one order of magnitude and pipe leak probabilities by two orders of magnitude approximately.

Evaluation

This research provides strong technical support for the NRC Piping Review Committee's non-mandatory recommendations aimed at reducing the number of snubbers at nuclear power plants. Flexible piping provides easier access for plant maintenance, reduces radiation exposures during maintenance, reduces thermal stresses during plant operation and positively affects construction and maintenance economics. Moreover, in general, piping reliability is improved.

Additional information on this work may be obtained from John O'Brien (x37854) of the Mechanical/Structural Engineering Branch, DET.

/S/

Robert B. Minogue, Director
Office of Nuclear Regulatory Research

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