✓
NUREG/CR-2877
SAND81-2613
RV
Printed August 1982

NUREG/CR--2877

DE83 000817

Investigation of Cable Deterioration in the Containment Building of the Savannah River Nuclear Reactor

Kenneth T. Gillen, Roger L. Clough, Lowell H. Jones

This report documents a part of the Qualification Testing Evaluation (QTE)
Program being conducted by Sandia National Laboratories.

Prepared by
Sandia National Laboratories
Albuquerque, New Mexico 87185 and Livermore, California 94550
for the United States Department of Energy
under Contract DE-AC04-76DP00789

Prepared for
U. S. NUCLEAR REGULATORY COMMISSION

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

NOTICE

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, or any of their employees, makes any warranty, expressed or implied, or assumes any legal liability or responsibility for any third party's use, or the results of such use, of any information, apparatus product or process disclosed in this report, or represents that its use by such third party would not infringe privately owned rights.

Available from
GPO Sales Program
Division of Technical Information and Document Control
U.S. Nuclear Regulatory Commission
Washington, D.C. 20555

and

National Technical Information Service
Springfield, Virginia 22161

NUREG/CR-2877
SAND81-2613
RV

Printed August 1982

MASTER

INVESTIGATION OF CABLE DETERIORATION
IN THE CONTAINMENT BUILDING
OF THE SAVANNAH RIVER NUCLEAR REACTOR

K. T. Gillen, R. L. Clough, and L. H. Jones

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

Sandia National Laboratories
Albuquerque, New Mexico 87185
operated by
Sandia Corporation
for the
U. S. Department of Energy

Prepared for
Electrical Engineering Branch
Division of Engineering Technology
Office of Water Reactor Safety Research
U. S. Nuclear Regulatory Commission
Washington, DC 20555
Under Interagency Agreement DOE 440-550-75
NRC FIN No. A-1051

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

ABSTRACT

This report describes an investigation of the deterioration of polyethylene and polyvinylchloride cable materials which occurred in the containment building of the Savannah River nuclear reactor located at Aiken, South Carolina. Radiation dosimetry and temperature mapping data of the containment area indicated that the maximum dose experienced by the cable materials was only 2.5 Mrad at an average operating temperature of 43°C. Considering this relatively moderate environment, the amount of material degradation seemed surprising.

To understand these findings, we undertook an experimental program on the commercial polyethylene and polyvinylchloride materials used at the plant to investigate their degradation behavior under combined γ -radiation and elevated temperature conditions. Several important aging effects were observed, including 1) a strong synergism between radiation and temperature, 2) large dose-rate dependent effects which occur over a wide range of dose rates, and 3) a dependence of the degradation in sequential radiation, elevated-temperature experiments on the ordering of the sequential exposure. The aging effects are discussed in terms of a chemical mechanism involving thermal breakdown of peroxides formed in reactions initiated by the radiation. Evidence for this mechanism is derived from infrared and chemiluminescence measurements and from chemical techniques.

It is established that the material deterioration at the plant resulted from radiation-induced oxidation and that the degradation rate can be correlated with local levels of radiation intensity in the containment area. It is concluded that test methods for aging and qualification testing of organic materials for use in a nuclear-plant environment should be designed to test for dose-rate effects and synergisms.

ACKNOWLEDGEMENTS

The authors are grateful for cooperation by Jim Leeth and other members of the staff at Savannah River who performed all of the temperature and dose-rate measurements inside the plant, in addition to providing cable samples, blueprints, and other pertinent information. The authors also acknowledge the assistance of Dave Haaland of Sandia who provided the FTIR spectra, and of John Nixon of Battelle Columbus who provided the chemiluminescence measurements. In addition the authors thank Ed Salazar, who served as a liaison with Savannah River staff during the early stages of this investigation and Carlos Quintana, who helped design and carry out a number of the experiments.

CONTENTS

	<u>Page</u>
INTRODUCTION	1
EXPERIMENTAL	1
Materials	1
Tensile Measurements	2
Infrared Spectroscopy	2
Mandrel Bend Tests	2
Chemiluminescence Measurements	3
PH ₃ Treatment of Polymer Samples	3
Radiation Aging Facility	3
Thermal Aging Facility	7
RESULTS AND DISCUSSION	10
Degraded Cable from the Reactor Containment Building	10
Environmental Mapping Inside the Containment Building	11
Accelerated Aging Studies	14
Chemical Mechanism of Degradation	21
SUMMARY AND CONCLUSIONS	27
REFERENCES	29

LIST OF TABLES

<u>Table</u>		<u>Page</u>
I	Sequential Experiments: Effect of PH_3	24
II	Sequential Experiments: Air versus Nitrogen	25

LIST OF FIGURES

<u>Figure</u>		<u>Page</u>
1	Cross Section of Cable	1
2	Radiation Aging Facility	4
3	High Dose-Rate Radiation Source	5
4	Low Dose-Rate Radiation Source	5
5	Radiation Test Chamber	6
6	Schematic View of Radiation Facility	8
7	Oven Aging Cell	9
8	Cross-Section of Reactor Building	10
9	Schematic Indicating Cable Location	11
10	Mechanical Properties versus Cable Position	12
11	Aging Results for PE in Air	15
12	Aging Results for PVC in Air	15
13	Aging Results for PE: Air versus Nitrogen	16
14	Aging Results for PVC: Air versus Nitrogen	16
15	Dose-Rate Effects for PVC	17
16	Dose-Rate Effects for PE	17
17	Thermal Aging of PE: Effect of Preirradiation	19
18	Thermal Aging of PVC: Effect of Preirradiation	20
19	Infrared Comparisons of Reactor Aged and Laboratory Aged PE	22
20	Chemical Mechanism of Radiation-Induced Oxidation	23
21	Effect of PH ₃ Treatment on Infrared Results for PE	24
22	Chemiluminescence Results for PVC	26

INTRODUCTION

Organic materials play an integral role in safety-related equipment inside the containment buildings of nuclear reactors. The largest single nuclear application of organic material is in cable insulation and jacketing. Seals, o-rings and gaskets, which are found in many types of equipment including motors, pumps, actuators, valves, transmitters and switches, are a second major application. Other examples include connectors, motor windings and varnishes, circuit boards, terminal blocks and lubricants. The aging behavior of the organic materials used in the containment area is an important engineering concern in nuclear power plant design.

In November, 1976, an inspection of a pump suction valve in the Savannah River K-reactor at Aiken, South Carolina, revealed badly embrittled polyethylene (PE) insulation on the power control wiring. A subsequent check of other cables in the K reactor, as well as in the similarly configured C and P reactors, brought to light additional sections of cable having embrittled PE. We undertook an investigation of the Savannah River cable materials with two main objectives:

- 1) to understand the cause of the degradation
- 2) to determine whether carefully designed aging tests would have indicated a potential problem with the use of this cable in the containment environment.

EXPERIMENTAL

Materials

We obtained a 60 m length of seven-conductor cable that had been removed from the C reactor. The cabling consisted of seven multi-stranded copper conductors each covered with a different colored polyethylene insulation (black, green, red, blue, orange, white, white with black stripes) and a thin nylon sleeve, all of which were enclosed in a polyvinyl chloride (PVC) jacket (Fig. 1). It had been purchased in

Figure 1. Cross section of cabling removed from the reactor containment building.

June of 1963 and had experienced an estimated 12 years of integrated reactor operation. The insulation was characterized as low to medium density polyethylene; the jacketing consisted of a carbon black filled PVC formulation. This type of PVC-jacketed cabling having PE insulation or crosslinked PE insulation was widely used in nuclear reactors built prior to the mid 1970's (1). For the laboratory aging experiments described in this paper, we were able to obtain cable samples from the Savannah River plant which had been purchased from the same manufacturer at the same time as the degraded cable in containment, but which had been kept at ambient temperature and had not been exposed to radiation. This material was of the same type as that removed from containment except that it was three-conductor (red, white and black PE) instead of seven-conductor cabling.

Tensile Measurements

Tensile tests were performed using an Instron Model 1130 Testing Machine equipped with pneumatic grips and having an extensometer clamped to the sample. After carefully removing both the wire conductor and the thin nylon sleeve, the PE samples were tensile tested as hollow tubes having 1 mm wall thickness. The PVC outer jacket was cut with a die into rectangular strip tensile samples having a 5.6 mm width. Samples were strained at 12.7 cm/min; initial jaw separation was 5.1 cm. The tensile test data given throughout this report represent the average obtained from three or more samples. The typical scatter in the data was about $\pm 20\%$ of the measured value in the case of the PE, and about $\pm 7\%$ of the measured value in the case of the PVC.

Infrared Spectroscopy

Infrared spectra were obtained with a Nicolet Model 7199 FTIR spectrometer. Samples were prepared by grinding the polymer at -196°C , mixing the resulting powder with ground KBr and pressure forming a pellet.

Mandrel Bend Tests

Mandrel bend tests were performed by wrapping the PE-insulated wires around a series of cylinders having progressively smaller diameters, and rating each test on a pass/fail basis according to a visual inspection for the appearance of cracks in the PE. Mandrel diameters used ranged from 40X down to 1X the diameter of the PE-insulated wire.

Chemiluminescence Measurements

Thermally induced chemiluminescence measurements were carried out on aged and unaged PVC samples by John Nixon of Battelle Columbus Laboratories using instrumentation similar to that described before (2). Samples were heated at 150°C with a slow, constant air flow. Light emission was monitored as a function of heating time.

PH₃ Treatment of Polymer Samples

PH₃ experiments were performed using a Parr bomb. Preirradiated samples were treated with 1.4×10^6 Pa (200 psi) of PH₃ for 24 hr at room temperature. The samples were then left standing open to the air for three days before elevated temperature aging.

Radiation Aging Facility

Figure 2 shows an artist's rendition of the radiation facility. Approximately 16,000 curies of Co-60 is positioned at the bottom of a water-filled stainless-steel lined concrete tank whose dimensions are 1.2 m by 2.4 m by 4.6 m deep. Radiation aging is carried out in water-tight test cells by lowering the cells to the bottom of the tank. Four meters of water, separating the Co-60 from experimenters at the top of the tank, provides radiation shielding. A water level control system and various radiation level monitors connected to an alarm system provide backup safety.

Details of the radiation aging portion of the facility are shown in Figs. 3 and 4. Two different spatial arrangements of Co-60 pencils exist, one for high dose-rate experiments, the other for intermediate to low dose-rate exposures. Figure 3 shows the arrangement used for high dose-rate exposures; it contains approximately 10,000 curies distributed in 12 cobalt pencils (dimensions of 1.14 cm diameter by 32 cm length) evenly spaced in a cylindrical holder of 18 cm diameter. Aging chambers can be placed either inside the cylinder of cobalt at the highest dose rate (~900 kR/hr) or in two positions adjacent to the cobalt cylinder. This cylindrical cobalt holder is supported at the end of two parallel channels. These channels also support the low dose-rate linear Co-60 holder and its associated air tanks and test cell holders, shown in Fig. 4. The cobalt holder for this linear array is configured similar to a test tube rack in which roughly 6,000 curies of cobalt-60 contained in 25 pencils is distributed approximately uniformly along its length. Test cell holders, each containing four cylindrical holes, are oriented parallel to the linear cobalt holder and are located on both sides of the source at various distances from it. Test cells placed in any of the holes of a given holder receive comparable radiation dose rates.

Figure 2. General view of radiation aging facility.

CIRCULAR ARRAY RADIATION SOURCE

Figure 3. Diagram of the high dose-rate portion of the aging facility.

LINEAR ARRAY RADIATION SOURCE

Figure 4. Diagram of the low dose-rate portion of the aging facility.

The particular dose rate is governed by the distance from the cobalt array and by the shielding between the test cell and the array. Gradients in dose rates occur in the individual test cells; for example, there is a dropoff in dose rates between the parts of the cell closest to, and furthest from, the cobalt. Minimizing these radial gradients requires moving the cell as far from the cobalt as possible. At the same time, it is desirable to minimize the amount of Co-60 required for a given dose rate at a given distance; this necessitates a minimum of shielding between the test cells and the cobalt. These two requirements were satisfied by using air tanks for filling any space between the Co-60 holder and the

test cell holders and by designing air-filled (water-tight) test cell holders. This arrangement not only minimizes gradients across the test cells for a given dose rate, but also minimizes the steps in dose rate occurring among adjacent test cell holders.

Another feature of this arrangement is the flexibility to spatially reposition the test cell holders in order to select appropriate dose rates for a given experiment. This also implies that new test cell holders capable of holding larger cells could easily be incorporated into the apparatus.

A detailed sketch of a test cell is shown in Fig. 5. The cylindrical sample aging region (10 cm diameter and 18 cm long) is located inside a brass can, which is in turn suspended from the

Figure 5. Underwater radiation test chamber.

lid of a double-walled stainless-steel can. (The double-walled design provides some thermal insulation.) When the stainless-steel lid is lifted free of the stainless-steel can, a brass lid at the bottom of the brass can is easily removed, allowing access to the sample aging region. The sample region is heated using an insulated nichrome wire wrapped around the sides of the brass can and also around a pancake heater which lies between the brass can and the stainless-steel lid. The auxiliary pancake heater was found to be useful for reducing rather substantial temperature gradients at the top of the sample aging region caused by the heat-sink effect of the massive top of the stainless-steel can. To accurately control and monitor the temperature in the sample region, two resistance temperature devices (RTDs) were incorporated in the design. A control RTD is directly clamped to the inside wall of the brass can and a monitor RTD is positioned close to the center of the sample-aging region. A 2.9 cm OD plastic tube runs from the top of the stainless-steel can up through the water tank. Lead wires for the heater and the two RTDs feed through this tube. In addition, a small tube inside the outer plastic tube is used to circulate air or other gaseous environments past the sample region at controlled flow rates. The temperature capabilities of the test cells are room temperature to 150°C. At 150°C, long term stability and accuracy of the temperature was found to be better than $\pm 0.3^\circ\text{C}$. Measured temperature gradients across the sample-aging region were less than $\pm 1.0^\circ\text{C}$.

Figure 6 shows the current arrangement of the aging facility viewed from the top of the tank. The cylindrical holder is positioned at one end of the tank. For the linear source, three adjacent test cell holders are located as close as possible to one side of the cobalt source. On the other side, a 23 cm wide air tank separates three adjacent test cell holders from the cobalt. Radiation dose rates at numerous locations in each aging position were measured using both a Victoreen Model 550 Radocon III Integration/Rate Electrometer and thermoluminescent CaF_2 wafers; agreement between the two methods was excellent. Since vertical and radial gradients exist in each aging location, sample positions during aging are always noted so accurate estimates of dose rates can be made. Quoted dose rates have an estimated uncertainty of $\pm 7\%$. Figure 6 lists the approximate center-of-can dose rates in the eight facility aging levels on January 1, 1981.

Thermal Aging Facility

Thermal aging is normally carried out in air-circulating ovens. There are numerous disadvantages and potential problems associated with this arrangement. Large temperature gradients occur in many ovens, and rapid movement of air past samples will often hasten the removal of volatile components (plasticizers, anti-oxidants), leading to unrealistically accelerated degradations. If two or more materials are aged in the same oven there is the danger that

Figure 6. Overhead schematic view of radiation aging facility showing center-of-can dose rate levels as of January 1, 1981.

outgassing or volatile degradation products from one material will affect the deterioration of another material. If, for this reason, only one material is aged in a given oven, the number of ovens required for a long-term program can be prohibitive.

With the above problems in mind, a unique and versatile thermal aging facility was constructed. The facility uses air-circulating ovens, each modified to accommodate a number of self-contained aging cells. A detailed sketch of one of these aging cells is shown in Fig. 7. It consists of a bell jar glass chamber which rests on a silicone gasket which in turn rests on an aluminum stand. A gas inlet line enters the sample aging region from the bottom through holes in the aluminum base and the silicone gasket. This arrangement allows aging to be carried out under various atmospheres (e.g., air, inert gas) and controlled gas-flow conditions. A small hole at the top of the glass chamber serves as a gas exit and as a means of introducing a permanently positioned thermcouple into the center of the sample region. Lead and aluminum collars help to insure an air-tight seal around the base of the glass chamber. These collars also improve the thermal

Figure 7. Oven aging cell.

stability and reduce temperature gradients in the sample aging region. A perforated stainless-steel sample basket sits inside of the glass chamber. To gain access to the samples, the glass chamber is lifted and tipped sideways, allowing the sample basket to slip free.

The aging cells are constructed in two sizes; smaller cells hold 5 cm ID by 13 cm-long sample baskets, while larger cells accommodate 7.5 cm ID by 13 cm-long baskets. Three of the smaller aging cells are incorporated in 27 liter air-circulating ovens. As many as six of the large cells have been placed in bigger ovens. Extensive temperature mapping inside the cells indicates that variations in temperature throughout the sample region are less than $\pm 1^{\circ}\text{C}$ at 150°C , and that much less short-term cyclic temperature fluctuation occurs inside a cell than in the surrounding oven.

RESULTS AND DISCUSSION

Degraded Cable From The Reactor Containment Building

On visual inspection, the 60 m cable removed from the Savannah River reactor building showed alternating areas of flexible and badly embrittled PE insulation. After removal of the PVC jacketing it was found that for the most severely affected regions, the extent of damage was such that handling or bending caused the PE insulation to crack and fall off of the wires. The installation position of the cable is indicated in Figs. 8 and 9 which are simplified diagrams of the reactor building. Near the -6 m (-20 ft) level the cable enters the cable tray at the point marked B in Fig. 9 and exits from containment at point M. Mandrel bend tests were performed on the PE insulation at a series of intervals along the cable. The upper plot in Fig. 10 gives the mandrel bend results for the cable as a function of position in the reactor (B-M refers to positioning in Fig. 9). A pronounced color dependency is evident, with radiation tolerance decreasing in the order: black > green > blue > red > orange, white, white with black stripes. The PVC jacketing, which showed no signs of visible degradation, was subjected to tensile elongation testing. The lower plot in Fig. 10 gives reduced elongation (e/e_0 , the experimental elongation divided by the value for PVC material in an unaged state), again as a function of position in containment. The areas of PVC showing reduced elongation correspond to areas having degraded PE. In areas where PE showed no evidence of degradation, the elongation data for PVC were essentially identical to values for unaged cable.

Figure 8. Cross sectional diagram of the reactor building.

Figure 9. Diagram of the containment building near the -6 m (-20 foot) level. The cable originates at a lower point A (not shown), enters the cable tray at B, and exits from containment at M. Numbers in boxes indicate measured dose rate values in rads/hr. Roman numerals in circles indicate the positioning of temperature monitors.

Elongation measurements were not taken on the PE due to experimental difficulties in stripping away the nylon sleeve, which tended to adhere strongly in the case of degraded specimens.

Environmental Mapping Inside the Containment Building

Radiation dosimetry mapping was carried out by Savannah River personnel at a series of points inside the reactor using temperature-luminescence dosimeters. Some of their results are shown in Fig. 9 where dose rates in rads per hour are

Figure 10. Mechanical properties versus cable position (B through M in Fig. 9). Upper plot: Mandrel bend results for PE insulation. Lower plot: Tensile elongation, e , for the PVC jacketing divided by the elongation value of unaged material.

indicated in boxes.*

*The literature is essentially devoid of dose rate measurements inside of reactor containment buildings, despite the importance of such data to predicting the long term aging behavior of organic materials in the containment area. Some theoretical calculations have been made (3).

Accurate dose rate measurements inside containment are in fact difficult. The values here are approximate, and probably represent lower limit values due to several factors: 1) since the temperature averaged about 43°C , a partial induced luminescence is expected to occur in the CaF_2 chips during the course of the measurement, thus reducing the apparent luminescence during the subsequent counting period; and 2) the chips were placed at the top of the cable trays, and were thus partially shielded by the cables from the D_2O lines below. Additionally, radiation levels are expected to fluctuate at a given point, due to variation in operation power, corrosion buildup and the speed of circulation of D_2O coolant. In particular, since the half lives of ^{16}N and ^{19}O in the cool water are very short, variations in coolant circulation rate could have an influence on radiation levels at positions near the coolant lines.

While dose rate measurements were unfortunately not made along the line of points B-D, measurements (Fig. 9) along the opposite side of the room are indicative of the relative magnitudes expected at B-D (values are not expected to correlate exactly, since the reactor is not equidistant from the two cable trays). Even with these small uncertainties, it is clear from Figs. 9 and 10 that the areas showing relatively higher damage of both PVC and PE correspond to areas of higher radiation dose rate, so that radiation is definitely implicated in the degradation process. The higher dose-rate areas result from proximity to 61 cm diameter pipes carrying D₂O coolant from the core to the heat exchangers, which pass 1.2 m under the cable trays. The radioactivity in the lines results from dissolved fission products, which have leaked into the coolant stream, as well as from at least two unstable isotopes, ¹⁹O and ¹⁶N, generated by irradiation of the D₂O:

In addition, Savannah River personnel monitored the ambient temperature at three locations (see points I-III in Fig. 9) inside the containment building during a period of ten months. The average temperature during operation was approximately 43°C. It varied between summer and winter and also varied as a function of power level of the reactor; the range was about 32° - 47°C. There was relatively little variation (less than $\pm 1^\circ\text{C}$) between the different measurement points.

The amount of degradation observed for a maximum estimated 12 year dose of only 2.5 Mrad is unexpected based on survey studies which purport to indicate typical radiation resistance of polymeric materials (4-6). For instance, a major compilation of radiation aging data is found in the report by the European Organization for Nuclear Research (CERN) entitled, "Selection Guide to Organic Materials for Nuclear Engineering"(4). The tables in this report indicate that at room temperature, radiation damage to polyethylene should be "incipient to mild" up to about 20 Mrad, "mild to moderate" up to about 90 Mrad, and "moderate to severe" up to about 400 Mrad. PVC is shown as exhibiting "incipient to mild" damage up to 10 Mrad, "mild to moderate" damage up to about 80 Mrad, and "moderate to severe" damage up to about 400 Mrad. In addition, another table (Table III.3) of this document indicates no major changes in radiation stability of these materials for radiation exposure at 85°C.

Accelerated Aging Studies

In an effort to replicate and understand the degradation of PE and PVC as seen in the plant, laboratory aging experiments were carried out on the "unaged" Savannah River cable under a variety of thermal, radiation and atmospheric conditions. We set out to conduct survey experiments using dose rates varying from about 1 Mrad/hr down to several krad/hr. The upper dose rate is typical of that used in material qualification testing evaluation programs (1); the lower limit was arrived at due to the practical decision of limiting experimental time periods to several months. Moderately elevated temperatures for survey experiments were chosen so as to slightly accelerate the degradation rates without exceeding temperatures which by themselves would cause significant material deterioration during the time scales of the experiments.

One of the first questions experimentally addressed was whether the degradation of the PE and PVC materials at Savannah River was due to specific interactions between the 4 cable components (copper, PE, PVC, nylon). For example, the polyethylene deterioration might have been accelerated by HCl produced as a result of irradiation of the PVC. For this reason, combined environment radiation at elevated temperature experiments in air (80°C, ~ 5 krad/hr) were carried out on: 1) complete, intact cable sections; 2) nylon-jacketed PE containing the copper conductor, but with the PVC removed; 3) PE containing the copper, but with nylon jacketing and PVC removed, and 4) PE that had been stripped from the copper conductor, and which had nylon and PVC coverings removed. Experiments were also performed with PVC jacketing on intact cables as well as with PVC that had been separated from the cable. Although substantial degradation was observed in each case based on tensile measurements, no significant differences in degradation rates were found for the PE and PVC materials when aged as separated components as opposed to intact cable sections.*

Given the above results, coupled with the desire to facilitate the experiments, the majority of the remaining studies were carried out on "prestripped" PE and PVC materials. Some striking observations from these aging experiments are clear from representative results shown in Figs. 11-16. Figure 11 is a plot of tensile elongation for PE as a function of aging time in three different environments: 1) elevated temperature

*However, this does not necessarily rule out a possible effect of copper on the degradation rate of PE, since PE samples stripped of the conductor may retain traces of copper contamination. Workers at Bell Labs have provided evidence of a strong degradation-promoting effect of copper conductor in the thermo-oxidative degradation of PE (7).

Figure 11. Tensile elongation results for PE insulation as a function of aging time in three different laboratory environments: 1) elevated temperature, 80°C; 2) radiation at room temperature, 5 krad/hr at 25°C; 3) radiation at elevated temperature, 5 krad/hr at 80°C.

Figure 12. Tensile elongation results for PVC jacketing material as a function of aging time in three different laboratory environments: 1) elevated temperature, 80°C; 2) radiation at room temperature, 4.4 krad/hr at 25°C; 3) radiation at elevated temperature, 4.4 krad/hr at 80°C.

Figure 13. Tensile elongation results for PE insulation as a function of aging time at 5 krad/hr, 80°C in atmospheres of air and of N_2 .

Figure 14. Tensile elongation results for PVC jacketing as a function of aging time at 4.4 krad/hr, 80°C in atmosphere of air and of N_2 .

Figure 15. Tensile elongation results for PVC jacketing as a function of dose at 43°C using three different dose rates.

Figure 16. Tensile elongation results for PE insulation as a function of dose at 43°C using three different dose rates.

only (80°C); 2) irradiation at room temperature (5 krad/hr; 25°C); 3) irradiation at elevated temperature (5 krad/hr; 80°C). The data shown is the average for the three colors of insulation tested (white, red, black). These results demonstrate the existence of a strong synergism between radiation and elevated temperature environments.

Aging data on the PVC jacketing material showed similar behavior. Plots of elongation vs. aging time under sets of environmental conditions similar to those used for the PE

are shown in Fig. 12. Again, important synergistic effects of radiation and moderately elevated temperature are observed. The above results are in marked contrast to the CERN data (4,5) which indicate radiation stabilities (mechanical) of 50 Mrad and 250 Mrad respectively for PVC and PE at 85°C.

Additional aging experiments in combined radiation-elevated-temperature environments were performed under an inert atmosphere of nitrogen. It was found that the exclusion of oxygen stopped the rapid synergistic degradation which would otherwise ensue. This is illustrated in Figs. 13 and 14 which show degradation data for PE and PVC under experimental conditions that differed only in the atmosphere used (air vs. N₂).

Figure 15 shows plots of tensile elongation for PVC as a function of total radiation dose for a series of combined environment exposures at 43°C and various radiation dose rates. A strong dose-rate dependency is obvious, with progressively higher damage-to-dose ratios obtained as the dose rate is lowered. PE was found to exhibit an even stronger dose-rate dependency trend (Fig. 16) with the extent of degradation at equivalent absorbed dose increasing, on moving to lower dose rates, by about a factor of 10 over the dose-rate range investigated.

The data shown in Figs. 15 and 16 cover more than 2 orders of magnitude in dose rate. Considering the trends observed, it would be reasonable to expect, that at the dose rate present in the region of the cable tray in the containment building, which was some 2 1/2 orders of magnitude lower than the lowest dose rate used in our experiments, the degradation rate would be even higher. The substantial degradation observed in cabling removed from the containment area (which had absorbed 2.5 Mrads) is thus readily understandable in terms of strong dose-rate effects in these materials. These results are not unusual since our extensive experimental program has revealed dose-rate effects at room temperature for a large number of other common cable insulation and jacketing materials, including ethylene propylene rubber and crosslinked polyolefin insulations and chloroprene and chloro-sulfonated polyethylene jackets (8).

For testing and qualification of materials for application in multiple stress environments (such as radiation and elevated temperature), the use of sequential aging experiments has been suggested(9). This prompted us to carry out a series of sequential aging experiments on the PE and PVC cable materials employing a) radiation at room temperature (4.4 krad/hr for PVC, 5 krad/hr for PE, both at 25°C for 83 days), and b) elevated temperature (80°C, no irradiation for 83 days). Tensile elongation values obtained are as follows: For (red) PE, radiation followed by elevated temperature: $e/e_0 = .17 \pm .04$; elevated temperature followed by radiation: $e/e_0 = .72 \pm$

.07. For PVC, radiation followed by elevated temperature: $e/e_0 = .32 \pm .02$; elevated temperature followed by radiation: $e/e_0 = .68 \pm .04$. It is seen that degradation is strongly dependent on the ordering of the sequential exposures; irradiation at room temperature followed by elevated temperature exposure is substantially more damaging than the result obtained when identical experiments were performed in the opposite order.

This behavior indicates that room temperature irradiation of the materials sensitizes them to subsequent thermal degradation. This sensitization is further illustrated in Figs. 17 and 18 which compare tensile data obtained during the thermal portion of a sequential experiment (radiation followed by elevated temperature) with data for exposure at the same elevated temperature, but without pre-irradiation. It is seen that a rapid degradation having no induction period occurs upon exposure of pre-irradiated samples of either PE or PVC to an elevated temperature environment. No substantial amount of the opposite phenomena, namely elevated-temperature sensitization of the polymers to radiation-induced degradation, was found with these materials. (However, Japanese workers have recently found evidence for such thermal sensitization in certain other types of materials (10).)

Figure 17. Tensile elongation results for PE insulation as a function of aging time at 80°C. Upper curve: unirradiated material. Lower curve: material preirradiated at 5 krad/hr for 83 days at 25°C in air.

Figure 18. Tensile elongation results for PVC jacketing as a function of aging time at 80°C. Upper curve: unirradiated material. Lower curve: material preirradiated at 4.4 krad/hr for 83 days at 25°C in air.

Cable materials that were aged under laboratory conditions showed several degradation characteristics analogous to those seen with the deteriorated cable found in the containment building. The polyethylene degradation rate in the three-conductor cable used in the aging experiments had a color dependency, with black insulation more stable than red which, in turn, was more stable than white. The magnitude of the differences between the colors depended on the particular experimental conditions. The color differences parallel the trend found for the cable removed from the containment building. The relative stability of the black insulation likely results from the carbon black coloring agent, which can act as an antioxidant (11). Color dependency in thermo-oxidative degradation of PE has been reported before (12), and follows approximately the same order of stability found here for radiation-induced degradation. In the naturally-aged cable removed from containment, as well as in laboratory aged samples, the nylon jacketing exhibited progressive yellowing and embrittlement on aging - roughly in correlation with advancing embrittlement of the PE. However, no quantitative measurements of nylon degradation were made.

Infrared spectroscopy on laboratory aged PE samples revealed a correlation between the formation and growth of a band in the carbonyl region (Fig. 19) and the decrease in tensile elongation. This carbonyl band is a characteristic end product of oxidation; it is broad, with multiple maxima, as would be expected for a collection of carbonyl groups of different structure. Infrared spectra were also obtained on PE at intervals along the cable removed from containment. It was found that embrittled regions showed strong carbonyl peaks while undamaged regions exhibited no absorption in this region (Fig. 19). Moreover, the shapes of the carbonyl bands in the damaged sections from containment and in the laboratory aged specimens were very similar, indicating that the chemistry induced in the laboratory aging experiments was similar to that in the reactor environment. From the above results, it appears that the degradation occurring under accelerated aging conditions and plant conditions is qualitatively similar.

Chemical Mechanism of Degradation

Oxygen diffusion effects have been cited before as a factor that can lead to dose-rate effects (13,14), and this is believed to be a contributing factor in the present case. However, the evidence tends to indicate that in this case a predominant reason for the aging phenomena observed (synergism, ordering dependence in sequential experiments, radiation sensitization, and dose-rate effects) results from a mechanism involving the formation and subsequent breakdown of peroxides. This can be visualized in terms of a time-dependent thermal amplification of the radiation damage, which results from the breakdown of peroxides as a rate-limiting step in the radiation-initiated degradation. Figure 20 presents a simplified degradation scheme (15). Chemical bonds are broken under the influence of high-energy irradiation to produce free radicals that react with O_2 to yield degradation products that include peroxides. The formation and build-up of substantial concentrations of hydroperoxides on room-temperature irradiation of PE and PVC samples in air has been documented (16). Peroxides are thermally labile; cleavage produces radicals, which, in the presence of oxygen, lead to more degradation and more peroxides in a chain-branching mechanism.

Kinetics of hydroperoxide homolysis in solution have been extensively investigated (17-19). Activation energies vary over a broad range (17) and can be substantially lower than the theoretical value for unimolecular dissociation (17,20), dependent on peroxide concentration, solvent environment and the presence of trace impurities. In bulk polymer specimens, products formed from radical chain reactions should be relatively immobilized, thus giving rise to localized high concentrations of peroxides and other oxidation products. Numerous impurities, including metals, are typically contained in commercial polymers. Evidence of breakdown of hydroperoxides on a short timescale has been seen before in thermo-oxidative studies of low density PE at 110°C (21).

Figure 19. Fourier Transform Infrared Spectra of PE insulation. Curve 1: Unaged laboratory sample. Curve 2: Sample removed from containment position H (see Figs. 9 and 10) which exhibited no apparent deterioration. Curve 3: Sample removed from containment position F (see Figs. 9 and 10) which exhibited substantial embrittlement. Curve 4: Laboratory aged sample (42 krad/hr, at 90°C to 17.9 Mrad total dose). Note the presence of the carbonyl bands near 1700 cm^{-1} for the aged samples.

Figure 20. Chemical mechanism for radiation-induced oxidation.

Given the above, it is not unreasonable that thermal breakdown of peroxides proceeds at a moderate, rate-determining level for the modest temperatures and reasonably long time scales used in this study.

A chemical technique was developed to substantiate the role of the peroxide-mediated pathway in the aging effects observed. The technique is modeled on a solution method for reducing hydroperoxides to alcohols by treatment with triphenylphosphine or related derivatives (22). Because it was necessary to carry out tensile tests subsequent to the experiments, dissolution was precluded. Instead, intact polymer samples were treated with gaseous phosphine, PH_3 , with the expectation that it could diffuse into the matrix to destroy existing peroxides.

Results of experiments in which PVC and PE specimens were irradiated in air at room temperature, treated with PH_3 for 24 hr and finally placed in an elevated temperature air environment are given in Table I together with the results for samples similarly aged but untreated with PH_3 . The PH_3 -treated samples exhibited essentially no degradation during subsequent thermal exposure. As shown in Figure 21, infrared results indicated that thermal exposure of the preirradiated, PH_3 -treated samples resulted in no increase in carbonyl absorption, in marked contrast to

TABLE I

SEQUENTIAL (RADIATION FOLLOWED BY ELEVATED TEMPERATURE) EXPERIMENTS:
EFFECT OF INTERVENING PH₃ TREATMENT ON TENSILE ELONGATION

Experimental Conditions ^a	e/e_o^b	
	PVC	PE
Unaged material	1.0 $\pm$ 0.05	1.0 $\pm$ 0.1
γ	0.74 $\pm$ 0.04	0.71 $\pm$ 0.08
γ ; T	0.38 $\pm$ 0.03	0.03 $\pm$ 0.03
γ ; PH ₃ ; T	0.74 $\pm$ 0.04	0.75 $\pm$ 0.08

^a γ = gamma radiation - 5 krad/hr (for PE), 4.4 krad/hr for (PVC) at 25°C for 96 days in air. T = thermal exposure of 80°C for 25 days in air. PH₃ = 1.4 x 10⁶ Pa (200 psi) of PH₃ at 25°C for 1 day.

^bSample tensile elongation divided by initial elongation.

Figure 21. Fourier Transform Infrared Spectra on sequentially aged PE samples with and without PH₃ treatment: 1) unaged material; 2) radiation aging only; 3) radiation aging followed by elevated temperature aging; 4) radiation aging followed by PH₃ treatment followed by elevated temperature aging. In each experiment, radiation was 4.4 krad/hr at 25°C (total dose = 10 Mrad). Thermal treatment was 80°C for 25 days. PH₃ treatment was 1.4 x 10⁶ Pa (200 psi) at 25°C for 1 day.

results obtained in the absence of intervening PH_3 treatment. These results offer strong evidence for the importance of hydroperoxide breakdown to the observed dose-rate, synergistic and ordering effects.

To provide evidence that oxygen is involved in both steps of the sequential radiation-thermal degradation mechanism, sequential experiments were run in which samples were 1) γ -irradiated at room temperature under nitrogen, then thermally aged in air; 2) γ -irradiated at room temperature in air, then thermally aged under nitrogen. Table II presents tensile data from these two experiments along with data from sequential experiments performed in an air environment throughout. Consistent with the mechanism in Fig. 20, it is clear that the degradation in the sequential experiment does require the presence of oxygen for both steps.

Samples of PVC were also examined using the technique of thermally induced chemiluminescence. Light output as a function of time for samples heated at 150°C was monitored for specimens having three different aging histories: a) unaged material, b) material irradiated at 4.4 krad/hr for 83 days at room temperature c) material irradiated at 4.4 krad/hr for 83 days at room temperature followed by elevated temperature treatment of 80°C for 83 days. Thermal luminescence curves are shown in

TABLE II
TENSILE ELONGATION RESULTS FOR SEQUENTIAL AGING EXPERIMENTS
IN NITROGEN ENVIRONMENTS

Experimental Conditions ^a	e/e_o^b	
	PVC	PE
Unaged material	1.0 $\pm$ 0.05	1.0 $\pm$ 0.1
γ in air	0.80 $\pm$ 0.04	0.68 $\pm$ 0.09
γ in air; T in air	0.32 $\pm$ 0.02	0.17 $\pm$ 0.1
γ in N_2 ; T in air	1.02 $\pm$ 0.05	1.01 $\pm$ 0.1
γ in air; T in N_2	0.83 $\pm$ 0.04	0.81 $\pm$ 0.08

^a γ and T have the same meaning as in Table I.

^b Sample elongation divided by initial elongation.

Fig. 22. Values measured for the initial burst of light (in counts per second above the instrument background levels of 100 cps) are as follows:

a) 150 cps; b) 1450 cps; c) 400 cps

The chemiluminescence phenomenon in organic materials is believed to be associated with accumulated peroxidic moieties. The data obtained are thus consistent with the mechanistic picture of the build-up of peroxides during the irradiation. Examination of Fig. 18, showing thermally-induced mechanical degradation of preirradiated PVC, would indicate a time-dependent

Figure 22. Chemiluminescence analysis of PVC samples (each sample run in duplicate). Light intensity in counts per second is shown, as a function of time, for samples heated at 150°C: a) unaged material, b) material irradiated at 4.4 krad/hr for 83 days at room temperature, c) material irradiated at 4.4 krad/hr for 83 days at room temperature followed by elevated temperature treatment of 80°C for 83 days. Instrument background count was 100 cps.

degradation rate for the pre-irradiated samples, such that the initially high rate diminishes with time. Consistent with this, the luminescence data indicate a time-dependent decrease in peroxide content during thermal treatment of pre-irradiated samples. This is seen by comparison of the luminescence for samples b and c, and also by the time-dependent behavior of the luminescence emitted by sample b.

SUMMARY AND CONCLUSIONS

Based on our laboratory experiments and on the information gathered at Savannah River, we conclude that the embrittled PE and partially damaged PVC found in the reactor containment building resulted from oxidative degradation. The degradation was initiated by the action of high energy radiation and required the presence of atmospheric oxygen. The radiation dose rate inside the containment area was seen to vary significantly from one location to another, with extensive material degradation associated with areas of locally high radiation intensity. We find no evidence that material interactions between the PE insulation and PVC jacketing of the cable played any significant role. The degradation rate of the PE insulation material was found to be strongly dependent on the coloring agent used, with black having the highest stability. We have demonstrated that the cable degradation which occurred in the reactor can be simulated quite well on a relatively short time scale under laboratory conditions in which both the radiation and the elevated temperature conditions are intensified.

In the past, studies intending to survey the radiation stability of organic materials have typically been performed over short timescales at very high dose rates (4,5) (on the order of 1 Mrad/hr or even 10 Mrad/hr). It is clear from the results obtained here that such survey results may be totally inappropriate when selecting materials for low-dose-rate, long-term applications.

The high degree of degradation exhibited by the Savannah River cable at a total absorbed dose of only 2.5 Mrad during its 12 yr service life, which seemed surprising by comparison with published data on radiation stabilities of the polymers involved, can be understood in terms of a time dependent effect. Our aging experiments provide unmistakable indications that the materials used at Savannah River would be expected to exhibit major degradation problems under low-level, long-term radiation conditions. This is clearly seen by the dose-rate experiments summarized in Figs. 15 and 16. An additional indication of long-term problems comes from the radiation-elevated temperature experiments as shown in Figs. 11 and 12 (increased temperature raises the reaction rates of the degradation pathways involved and results in an acceleration of the time scale).

Current materials aging methodology used for nuclear qualification as given by IEEE (9) specify an upper limit of 1 Mrad/hr on the dose rate for radiation aging. Our work brings to light the occurrence of large dose-rate effects spanning several orders of magnitude of dose rates below this level. One approach that has been applied to accelerated radiation aging is an attempt to compensate for any dose-rate effects by increasing the total dose given in the aging experiment. One of the IEEE standards for nuclear qualification refers to two empirical formulas for estimating the "overdose" necessary (9a). Applying one of these formulas (23) to the present case (25 rads/hr maximum for 12 years giving an integrated dose of 2.5 Mrad in the reactor) it would be predicted that an overdose by a factor of 2.5 would be necessary to simulate the plant condition using a 1 Mrad/hr dose rate. This equates to a total dose of 6 Mrad. Examination of Figs. 15 and 16 reveal that after 6 Mrad at 1 Mrad/hr, the observable mechanical damage is negligible, in strong contrast to what was actually observed at the reactor. Applying the other of these formulas (24), an entirely different prediction is obtained, with a calculated overdose by a factor of 66, or 165 Mrad total dose. Ongoing experiments in our laboratories with other cable insulation types are revealing that the magnitude of dose-rate effects varies tremendously from one material type to another. The use of any empirical "overdose" approach for accelerated aging has serious drawbacks, in that the dose chosen may lead to a substantial underestimation of damage in the case of materials having very large dose-rate effects, while overestimating damage for materials having minor dose-rate effects.

Additionally, the IEEE guidelines (9) suggest the use of sequential experiments for laboratory simulation of multiple stress environments such as radiation and elevated temperature, and suggest a sequence involving thermal aging in an air circulating oven followed by radiation exposure (9b). The results obtained here demonstrate that in fact the ordering of such sequential exposures is an important consideration, since it can have a tremendous effect on the extent of degradation incurred by polymeric materials. Moreover, we find that for the materials investigated here, the correlation between sequential aging experiments and combined environment aging is not straightforward. Also, simple thermal aging data of polymers can become irrelevant when radiation is a factor, since the radiation can drastically alter the susceptibility of the material to thermal degradation.

This work illustrates how radiation-thermal aging tests using several dose rates and several temperatures can be useful to indicate in advance that major degradation problems would be expected with the long-term use of certain organic materials in a nuclear reactor containment application.

REFERENCES

1. Personal communication from W. Booth, USNRC, Nov. 20, 1981 and Mar. 17, 1982. Information taken from licensee responses to I and E Bulletin 79-01B, documenting "Environmental Qualification of Safety-Related Electrical Equipment."
2. G. D. Mendenhall, Angew. Chem. Int. Ed. Eng., 16, 225 (1977).
3. J. Sejvar, Nuclear Technology, 36, 48 (1977).
4. M. H. VandeVoorde and C. Restat, Selection Guide to Organic Materials for Nuclear Engineering, CERN 72-7, Geneva (1972).
5. H. Schonbacher and A. Stolarz-Izycka, Compilation of Radiation Damage Test Data, CERN 79-4, Geneva (1979).
6. R. W. King, N. J. Broadway and S. Palinchak, The Effect of Nuclear Radiation on Elastomeric and Plastic Components and Materials, REIC Report No. 21, Sept. 1, 1961.
- 7a. W. L. Hawkins, M. G. Chan and G. C. Link, Polym. Eng. Sci., 11, 377 (1971).
- b. H. M. Gilroy, Long-Term Photo- and Thermal Oxidation of Polyethylene, in R. B. Eby (Ed.), Durability of Macromolecular Materials, American Chemical Society, Washington, DC (1979) p. 62.
8. K. T. Gillen and R. L. Clough, Rad. Phys. and Chem., 18, 679 (1981).
- 9a. IEEE Standard for Qualifying Class 1E Equipment for Nuclear Power Generating Stations, IEEE Std. (323-1974), Institute of Electrical and Electronics Engineers, Inc., New York (1974).
- b. IEEE Standard for Type Test of Class 1E Electric Cables, Field Splices, and Connections for Nuclear Power Generating Stations, IEEE Std. (383-1974), Institute of Electrical and Electronic Engineers, Inc., New York (1974).
10. E. Oda, K. Uchida, S. Fujimura and S. Oya, Radiation Resistance of Insulation Materials for Electric Cables, EIM-76-38, 1976.
11. J. R. Shelton, "Stabilization Against Thermal Oxidation," in W. L. Hawkins (Ed.), Polymer Stabilization, Wiley, New York (1972) p. 64.
- 12a. J. B. Howard, Proc. 21st Int. Wire and Cable Symp., 329 (1972).
- b. H. M. Gilroy, unpublished results.
13. W. Schnabel, "Degradation by High Energy Irradiation." In H. H. G. Jellinek (Ed), Aspects of Degradation and Stabilization of Polymers, Elsevier, Amsterdam, 1978, Chap. 4.

14. I. Kuriyama, N. Hayakawa, Y. Nakase, J. Ogura, H. Yagyu and K. Kasai, IEEE Trans. Elect. Insul., EI-14, 272 (1979).
15. For more extensive descriptions of free radical oxidation reaction steps, see (a) J. Rabek, Oxidative Degradation of Polymers, in C. Bamford and C. Tipper, Eds., Comprehensive Chemical Kinetics, 14, Degradation of Polymers, Elsevier, Amsterdam, 1975, p. 425. (b) Y. Kamiyua and E. Niki, Oxidative Degradation in H. H. G. Jellinek, Ed., Aspects of Degradation and Stabilization of Polymers, Elsevier, Amsterdam, 1978, p. 79. (c) J. Shelton, Stabilization Against Thermal Oxidation, in L. Hawkins, Ed., Polymer Stabilization, Wiley, New York, 1972, p. 29. (d) M. Dole, The Radiation Chemistry of Macromolecules, Vol. II, Academic, New York, 1973, p. 263. (e) L. Reich and S. Stivala, Elements of Polymer Degradation, McGraw-Hill, New York, 1971.
- 16a. C. Decker, F. Mayo, and H. Richardson, J. Polym. Sci., Polym. Chem. Ed., 11, 2879 (1973).
- b. C. Decker, J. Appl. Polym. Sci., 30, 3336 (1976).
17. R. Hiatt, Hydroperoxides, in D. Swern, Ed., Organic Peroxides Vol. II Wiley, New York, 1971, p. 94.
18. J. Chien, Hydroperoxides in Degradation and Stabilization of Polymers in G. Geuskens, Ed., Degradation and Stabilization of Polymers, Halsted, New York, 1975, p. 95.
19. F. Tudos, T. Kellen, and T. Nagy, Thermo-oxidative Degradation of Poly(vinyl chloride), in N. Grassie, Ed., Developments in Polymer Degradation, Applied Science, Essex, England, 1979 p. 187.
20. S. W. Benson, J. Chem. Phys., 40, 1007 (1964).
21. H. N. Cheng, F. C. Schilling, and F. A. Bovey, Macromolecules, 9, 363 (1976).
22. R. Mair and R. Hall, Determination of Organic Peroxides, in I. Kolthoff and P. Elving, Eds., Treatise on Analytical Chemistry, Vol. 14, Pt. 2, Wiley, New York, 1971, p. 295.
23. ASTM Standard Classification System for Polymeric Materials for Service in Ionizing Radiation, ASTM D 2953-71.
24. IEEE Guide for Classifying Electrical Insulating Materials Exposed to Neutron and Gamma Radiation, IEEE Std. (278-1967), Institute of Electrical and Electronic Engineers, Inc., New York, (1974).

Do Not
Microfilm Distr.

DISTRIBUTION

U. S. Nuclear Regulatory Commission
Distribution Contractor (CDSI) (290 copies)
265 copies for RV
25 copies for NTIS
Division of Document Control
Distribution Services Branch
7300 Pearl Street
Bethesda, MD 20014

AERE Harwell
Didcot Oxfordshire
OX 11 0RA
England
Attn: D. Phillips

AMN Impianti Termici E. Nucleari
Via Pacinotti, 20
16151 Genova Sampierdarena
Italy
Attn: G. Bottinelli

AMN Spa
Via G. D'Annunzio 113
16121 Genova
Italy
Attn: P. Giordano

AMN Spa
Centro Sperimentale del
Boschetto
Corso F. M. Perrone, 118
16161 Genova
Italy
Attn: C. Bozzolo

ASEA-ATOM (3)
Engineering
Box 53
S-721 04
Vasteras
Sweden
Attn: G. Kvist
A. Kjellberg
B. Langser

ASEA KABEL AB
P. O. Box 42108
S-126 12
Stockholm
Sweden
Attn: C. T. Jacobsen

Atomic Energy of Canada, Ltd.
Chalk River
Ontario
Canada
Attn: G. F. Lynch

Atomic Energy of Canada, Ltd. (2)
Power Projects
Sheridan Park Research Comm.
Mississauga, Ontario L5K1B2
Canada
Attn: C. Byrne
P. Gumley

Babcock Brown Boveri Reaktor
Heppenheimer Strasse
6800 Mannheim 31
Postfoche 410323
West Germany
Attn: R. Schemmel

Battelle Institute V
Am Romerhof 35
D6000 Frankfurt/M
West Germany
Attn: B. Holzer

Belgonucleaire
rue de Champ-de Mars, 25
B-1050 Brussels
Belgium
Attn: J. P. Van Dievoet

Bhabha Atomic Research Center
Trombay, Bombay
400 085
India
Attn: S. K. Mehta

British Nuclear Fuels, Ltd.
Springfield Works
Salwick, Preston
Lancs
England
Attn: W. G. Cunliffe, Bldg 396

Bundesanstalt fur Material Prufung
Unter den Eichen 87
1 Berlin 45
West Germany
Attn: K. Wundrich

Centre d'Etudes Nucleaires de
Fontenay-aux-Roses (3)
BP No. 6
92260 Fontenay-aux-Roses
France
Attn: Cogne
M. Lemeur
G. Ganouna-Cohen

Centre d'Etudes Nucleaires de Saclay(2)
BP No. 21
91190 Gif-Sur-Yvette
France
Attn: G. Gaussens
J. Chenion

CERN
Laboratoire 1
CH-1211 Geneva 23
Switzerland
Attn: H. Schonbacher

Commissariat a l'Energie Atomique
CEN Cadarche
BP No. 1
13115 Saint Paul Lez Durance
France
Attn: J. Campan

Conductores Monterrey, S. A.
APDO Postal 2039
Monterrey, N. L.
Mexico
Attn: Ing. Patricio G.
G. Murga

Electra de Viesgo, S.A.
Departamento Nuclear
Medio, 12 - Santander
Spain
Attn: J. L. del Val

Electricite de France (5)
Service Etudes et Projects
Thermiques et Nucleaires
Tour E.D.F. - G.D.F. Cedex No. 8
92080 Paris la Defense
France
Attn: J. Roubault
M. Barbet
A. Janoir
Y. LeLouarn
J. Delaire

Electricite de France (3)
Direction des Etudes et Recherches
Les Renardières - Route de Sens
Eucluelles - 77250 Moret Sur Loring
France
Attn: G. Delmas
P. Roussaire
V. Deglon

Electricite de France (4)
DER
1 Avenue du General de Gaulle
92141 Clamart Cedex
France
Attn: J. Clade
A. Lacoste
J. Ribot
L. Deschamps

EURATOM
C.E.C. J.R.C.
Ispra (Varese)
Italy
Attn: G. Mancini

EURATOM
Joint Research Centre
Petten Establishment
European Communities
Petten
The Netherlands
Attn: M. Van de Voorde

Fisher Controls, Ltd.
Century Works
Lewisham
London, SE 137 LN
England
Attn: P. A. Graves

Framatome (2)
Tour Fait
Cedex 16
92084 Paris la Defense
Cedex 16
France
Attn: J. Meyer
G. Chauvin

Furukawa Electric Co., Ltd.
Hiratsuka Wire Works
1-9 Higashi Yawata 5 Chome
Hiratsuka, Kanagawa Pref.
Japan 254
Attn: E. Oda

Gesellschaft fur Reaktorsicherheit
Glockengasse 2
6000 Cologne 1
West Germany
Attn: E. Dluzniewski

Hitachi Cable
777 Third Avenue
New York, NY 10017
Attn: H. J. Amino
M. Sasson

Imatran Vioma OY
Power Station Department
Elrikinkatu 27
Box 138
00101 Helsinki 10
Finland
Attn: B. Regnell

Institute of Radiation Protection
Department of Reactor Safety
P.O. Box 268
00101 Helsinki 10
Finland
Attn: M. Keikkila

International Atomic Energy Agency
Wigramerstrasse 5
P.O. Box 100
A-1400 Vienna
Austria
Attn: S. Machi

ITT Cannon Electric Canada
Four Cannon Court
Whitley, Ontario, L1N5V8
Canada
Attn: B. Marshall

Japan Atomic Energy Research Institute
Takasaki Radiation Chemistry
Research Establish Establishment
Watanuki-Machi
Takasaki, Gunma-Ken
Japan
Attn: N. Tamura

Japan Atomic Energy Research Institute
Tokai-Mura
Naka-Gun
Ibaraki-Ken
Japan
Attn: A. Kohsaka

Kansai Electric Power Co., Inc.
1725 K Street NW
Suite 810
Washington, DC 20006
Attn: J. Yamaguchi

Kraftwerk Union Aktiengesellschaft (2)
Hammerbacherstrasse 12 + 14
D-8520 Erlangen
West Germany
Attn: W. Morrell
I. Terry

Meideusha Electric Mfg. Co., Ltd.
1-17, 2-Chome Osaki
Shinagawa-Ku
Tokyo
Japan
Attn: M. Kanazashi

Motor Columbus
CH-5401
Parkstrasse 27
Baden
Switzerland
Attn: H. Fuchs

NOK AG Baden
Beznau Nuclear Station
CH-5312 Doettingen
Switzerland
Attn: O. Tatti

Ontario Hydro
700 University Avenue
Toronto, Ontario, M5G1X6
Canada
Attn: R. Wong

Osaka Laboratory for Radiation Chemistry
Japan Atomic Energy Research Institute
25-1 Mii-manami Machi,
Neyagawa-shi
Osaka 572
Japan
Attn: Y. Nakase

Oy Stromberg Ab,
Helsinki Works
Box 118
SF-00101
Helsinki 10
Finland
Attn: P. Paloniemi

Rhein-Westf TUV
Stuebenstrasse 53
D-43 Essen
West Germany
Attn: R. Sarturi

Studsvik Energiteknik AB
S-61182
Nykoping
Sweden
Attn: E. Hellstrand

Sydskraft
Southern Sweden Power Supply
21701
Malmo
Sweden
Attn: O. Grondalen

Tokyo Electric Power Co., Inc.
No. 1-3 1-Chome Uchisaiwai-Cho
Chiyoda-Ku
Tokyo 100
Japan
Attn: J. Hamada

Traction & Electricite
Rue de la Science 31
1010 Brussels
Belgium
Attn: P. A. Dozinell

Universidade Federal de Rio de Janeiro
Av. N.S. Copacabana 661/1201
22050 Rio de Janeiro
Brazil
Attn: Paulo Fernando Melo

Waseda University
Department of Electrical Engineering
170-4 Shinjuku
Tokyo
Japan
Attn: K. Yahagi

Westinghouse Nuclear Europe (3)
Rue de Stalle 73
1180 Burssels
Belgium
Attn: R. Minguet
R. Doesema
J. Cremader

1000 J. K. Galt
1154 D. A. Bouchard
1200 G. Yonas
1214 A. Owyong
1214 A. V. Smith
1800 R. S. Claassen
1810 R. G. Kepler
1811 L. A. Harrah
1811 R. L. Clough
1813 J. G. Curro
1813 K. T. Gillen (10)
1815 R. T. Johnson
1815 S. R. Kurtz
2155 J. E. Gover
2155 O. M. Stuetzer
2341 M. B. Murphy
9200 W. C. Myre
9300 R. L. Peurifoy, Jr.
9400 A. W. Snyder
9410 D. J. McCloskey
9420 J. V. Walker
9440 D. A. Dahlgren
9441 M. Berman
9442 W. A. von Riesenmann
9443 D. D. Carlson
9444 S. L. Thompson
9445 L. O. Cropp
9445 C. M. Craft
9445 D. T. Furgal
9445 W. H. McCulloch
9445 F. J. Wyant
9445 J. S. Yu
9446 B. E. Bader
9446 L. L. Bonzon (15)
9446 W. H. Buckalew
9446 L. D. Bustard
9446 D. M. Jeppesen
9446 E. E. Minor
9446 E. A. Salazar
9446 F. V. Thome
9446 R. E. Trujillo
9450 J. A. Reuscher
9452 M. Aker
9452 J. Bryson
9452 D. McKeon
9452 J. S. Philbin
9700 E. H. Beckner
8214 M. A. Pound
3141 L. J. Ericksen (5)
3151 W. L. Garner (3)