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13. ABSTRACT (Maximum 200 words) Capacitor-grade polypropylene films were aged under multiple stresses (electrical, thermal, radiation) using a 2MW thermal nuclear reactor. Thermal and electrical stresses were found to slightly decrease both the AC and DC breakdown voltages. Radiation stresses were found to increase the breakdown voltages. The same trends were seen in volume resistivity. The radiation effects dominate both the thermal and electrical effects and are attributed to increased crystallinity of the polypropylene. However, mechanical properties are significantly degraded by radiation damage, and degree of breakdown enhancements are seen for combined stresses. Xomputer simulation has been performed showing the magnitude of the neutrine signal from a 100 kilocurie tritium source, under the geometrical conditions of the experiments at Los Angeles National Laboratory, TA-33, is about an order of magnitude smaller than the gravity signal from a 2600 gr mass (the assumed value for the mass of the source). The observations were performed by using Professor Joe Weber's torsion balance, a room-temperature instrument.				
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FINAL REPORT

December 1, 1990

**MULTIFACTOR STRESS AGING
OF ELECTRICAL INSULATION**

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RESEARCH OBJECTIVES

To establish a physical and experimental base in order to study and understand multifactor stress (radiation, thermal, electrical) aging of electrical insulation.

ABSTRACT

Capacitor-grade polypropylene films were aged under multiple stresses (electrical, thermal, radiation) at the University at Buffalo 2 MW Thermal Nuclear Reactor. The aging experiments were performed for singular stresses as well as a combination of any two stresses and finally for simultaneous application of all three. The radiation was applied to electrical insulation by suspending a capsule in one of the reactor's stand pipes. The capsule was exposed to combined neutron-gamma radiation with the thermal neutron flux rate of 2.2×10^{10} n/cm².s, fast neutron flux rate of 8.8×10^9 n/cm³.s and gamma radiation at a level of 1.5×10^6 rad/h. Polypropylene film 25.4 μ m thick was wound between two aluminum electrodes and then the ac voltage 1000 V_{rms} was applied, giving the electrical stress of 40 V_{rms}/ μ m. To thermally stress the electrical insulation, the capsule was wound with a heating filament giving temperature rise to 90 °C within the capsule in the 42 °C ambient temperature environment. After one hour aging experiment and two months radioactive cooling down, the films were unwound and diagnosed for changes in the electrical, mechanical and physico-chemical properties. The results obtained, and the characterization and identification of degradation and failure mechanism under multifactor stress (radiation, thermal, electrical) is reported in this Final Report.

I. INTRODUCTION

As technology advances, increasing demands are made on materials and components. The most important requirement is the reliable operation under various operating and environmental conditions. Radiation has been added to the growing list of combined environments which already include temperature, electric stress, pressure, atmosphere, humidity, acceleration, shock and vibration [1]. There is need, therefore, for materials and components to perform reliably under this multistress environment.

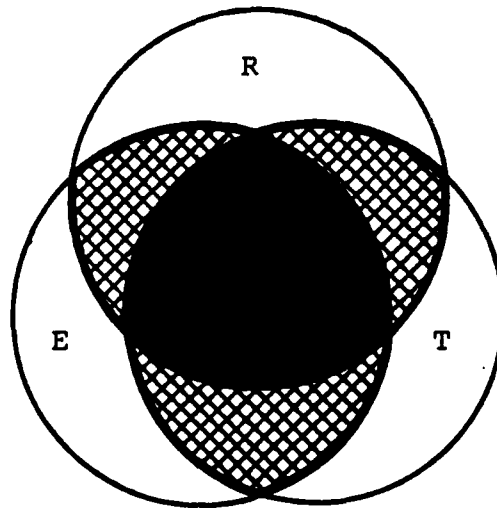
Nuclear sources for electric power generation are well established for earth-based applications. Nuclear power sources are also promising candidates for future space power. As electrical power requirements increase with each succeeding space program, new technologies and often new energy sources must be used to provide the optimum power system. The electrical power systems used in earlier space programs consisted of: primary batteries (non-rechargeable) - short duration, low power; chemical dynamic systems - short duration, high power; fuel cells - medium duration, medium power; radioisotope thermoelectric or dynamic systems - long duration, low power; and solar energy photovoltaic systems - low duration, medium power [2].

All of the above low or medium-level power systems are not applicable for prime power in future space programs. For long durations applications for which extremely high power levels (hundreds of kilowatts) are required, nuclear dynamic system is the most

potential candidate [2]. With the numerous opportunities that the nuclear power generation offers, there is also a unique problem of self-generated neutron and gamma radiation. The utilization of nuclear power as a source of energy for earth and space-based power sources provides, therefore, the designer with a radically different and challenging environment. All matter is affected to some extent by radiation. Organic materials along with semiconductors are most seriously affected by radiation, while metals and ceramics are, in general, more resistant to it. There is a significant number of electrical components and seals in nuclear reactor environments requiring polymeric materials in their design and construction. A few of these components include power cables, capacitors, transformers, control cables, instrumentation cables, pump and fan driving motors, valves, relays and connectors [3]. The most extensive use of organic polymers in a radiation environment is in the form of cable insulation inside the containment building of a nuclear power plant, where hundreds of miles of cable can be found, such as in the Waterford III reactor [4]. Another application of organic insulation inside the containment can be found in high voltage capacitors, where they protect the primary coolant motors from overvoltage surges [5]. Whatever the application, polymers and other organic materials are typically found among the weak links in equipment where radiation aging is a factor. This is especially true in the case of an oxygen containing atmosphere, where oxidative degradation can be induced by a number of different stresses including radiation,

thermal, electrical, photochemical and mechanical stresses [6]. Thermally induced aging is the most frequently encountered and studied aging stress. Predictions are considerably more difficult in environments where a stress other than thermal is dominant because aging methodology involving other stresses is less developed. In environments where two or more stresses are important, the situation is even more difficult [6]. A review on the influence of radiation and multistress aging on electrical insulation was included in the First Annual Report submitted to the AFOSR on December 15, 1989 [7], and is not repeated here. It concluded that no data is available on multistress (electrical, thermal, radiation) aging of electrical insulation, as is depicted graphically in Figure I.1.

Considering the importance of multifactor stress on electrical insulation, the objective of this work is to find how a combination of radiation, electrical and thermal stresses can affect the organic insulation materials. Main focus is placed on determination of changes in electrical, mechanical and morphological properties of polymer insulating films upon multifactor stress (radiation, thermal, electrical) aging. Polypropylene, being the primary organic dielectric used for capacitors located inside the containment, was chosen as a subject of this study. With a significant lack of information on the changes taking place in polymer insulation under influence of multistress aging, this is an important introductory work in the area of multifactor stress aging.



Data available



Some data available



No data available

Figure I-1 Multifactor stress aging of electrical insulation
(T: Thermal; E: Electrical; R: Radiation)

II. MULTIFACTOR STRESS AGING OF ELECTRICAL INSULATION

Capacitor-grade polypropylene films were aged under multiple stresses: electrical, thermal and radiation. The aging experiments were performed for singular stresses as well as a combination of any two stresses and finally for simultaneous application of all three. The detailed experimental procedure was described in the First Annual Report to the AFOSR [7], it is also described in the Appendix. In summary, the polypropylene film was exposed to combined neutron-gamma radiation with the thermal neutron flux rate of 2.2×10^{10} n/cm².s, fast neutron flux rate of 8.8×10^9 n/cm².s and gamma radiation at a level of 1.5×10^6 rad/h. The capacitor was also stressed to 1000 V_{rms}, 60 Hz voltage, giving the electric field of 40 V_{rms}/μm. To thermally stress the electrical insulation, the film was heated to 90 °C in the 42 °C ambient temperature environment. After two-month period of radioactive cooling down of the samples, the capacitors were unwound and stored for poststress characterization of the polypropylene film.

The effect of multifactor stress aging on electrical, mechanical and physico-chemical properties of capacitor grade polypropylene film were then studied in order to characterize the nature of changes and to identify degradation and failure mechanisms. In order to study the effects of multifactor stress-aging on polypropylene, a large number of experiments were designed and performed. These experiments consisted basically of five electrical, three mechanical and five morphological and chemical diagnoses. A flow chart of the various experiments performed is shown in Figure II-1.

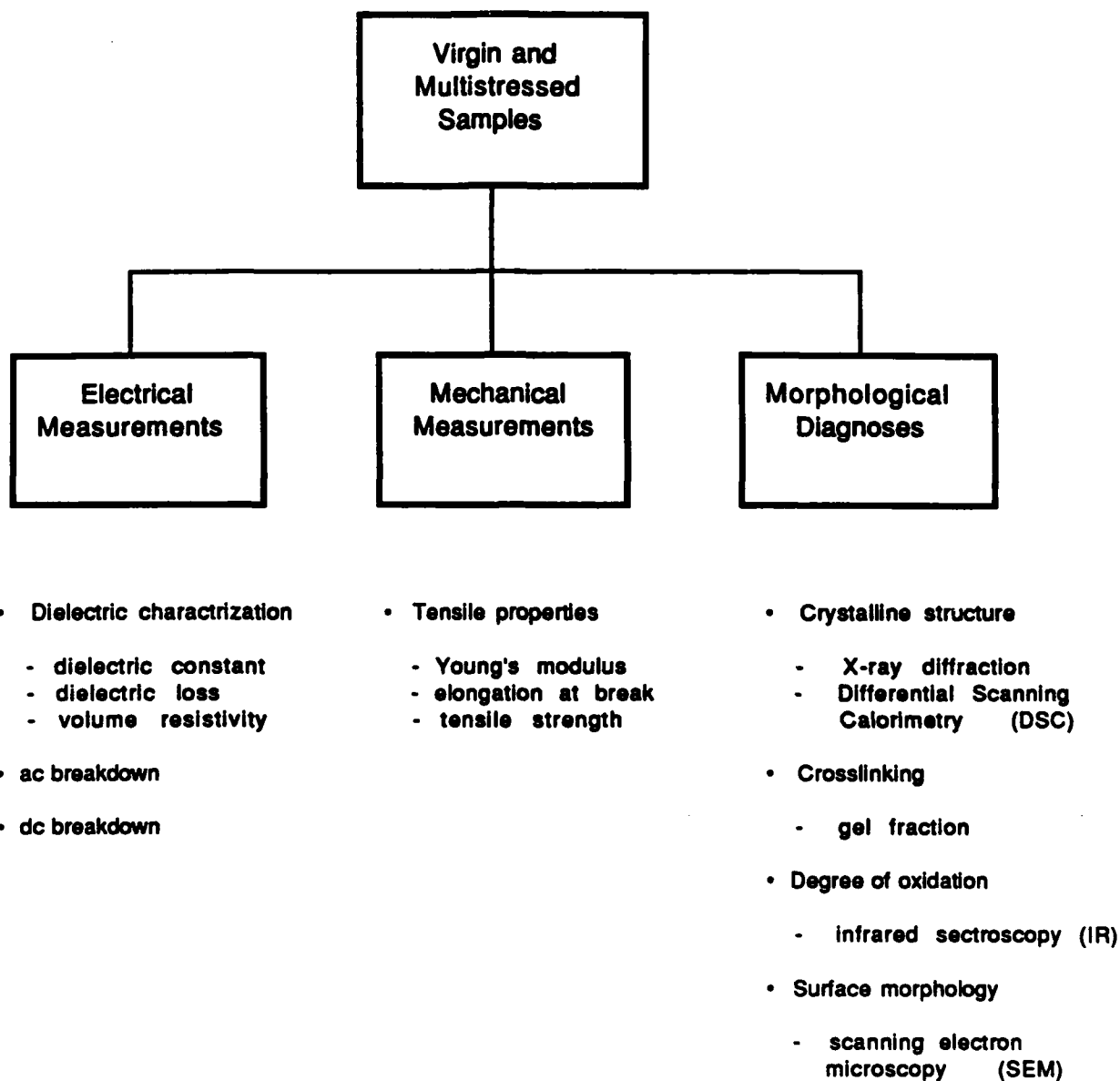


Figure II-1 Flow chart of experiments performed

The electrical measurements consisted of ac and dc breakdown voltage measurements and dielectric characterization, which include dielectric constant, dielectric loss and volume resistivity. The mechanical measurements included changes in tensile properties such as Young's modulus, elongation at break and tensile strength. The morphological and chemical diagnoses that were carried out included surface morphology (scanning electron microscope - SEM), crystalline structure (X-ray diffractometry and differential scanning calorimetry - DSC), crosslinking level (gel fraction), identification of chemical bonds and degree of oxidation (infrared spectroscopy).

All mentioned above measurements are of great importance in determination of polymer's stability under environmental stresses such as radiation, elevated temperature and electric field. For example, changes in the dielectric constant, dielectric loss and volume resistivity electrically characterize the polymer in terms of energy storage capability, efficiency and charge leakage tendency [98]. Measurements of the electrical breakdown strength determine the maximum withstand voltages. Tensile properties measured indicate the maximum ability of the material to withstand mechanical stresses without fracture, rupture or cracking. Mechanical measurements are important also from an electrical standpoint, because failure mechanism of the electrical insulator under influence of radiation or elevated temperature might be due to cracking and embrittlement, rather than homogeneous decay in dielectric strength [6]. Finally, physical and chemical properties measured yield information in identifying

the nature of the changes taking place in the polymer under influence of multifactor stresses.

The data obtained was analyzed and discussions are included in the Appendix, as a journal paper "Effects of Multistress Aging (Radiation, Thermal, Electrical) on Polypropylene;" submitted to the IEEE Transactions on Nuclear Science and recommended for publication after minor editorial changes (letter from the journal editor is also included). The conclusions obtained from this study follow in the next section.

III. CONCLUSIONS

The effect of multifactor stress aging on electrical, mechanical and physico-chemical properties of capacitor grade polypropylene film was studied in order to characterize the nature of changes and to identify degradation and failure mechanisms. Details of the experimental procedure and the results obtained are provided in the Appendix. The electrical properties measured comprised of dc/ac breakdown strengths, volume resistivity, as well as in-situ and post-stress dielectric properties.

Both thermal and electrical stresses decreased slightly the ac and dc breakdown voltages of polypropylene. The radiation stress, on the other hand, increased the breakdown strength of the polymer for both ac and dc electric field stresses. A similar trend was also observed for volume resistivity. The radiation stress is the most dominant factor influencing both the electric strength and the volume resistivity. The combination of either thermal or electrical stress with the radiation is overshadowed by the latter and overall increase in the above properties is observed. This increase is attributed to the increase in the degree of crystallinity of the polypropylene.

The most pronounced changes were observed, however, in the mechanical properties of the film. There is a significant decrease in elongation-at-break and tensile strength proving deterioration of the polypropylene under combined neutron-gamma radiation. This degradation is attributed to chain scission of the polypropylene molecules. The temperature stress had an opposite effect causing

an increase in the above mentioned properties and offsetting, therefore, the negative effect of radiation. In addition to the discussed changes, strong interactive (synergistic) effects are observed. The dc breakdown strength and the Young's modulus are much lower for combined E/T/R stresses than expected by merely adding the effects of each stress individually.

Based on this work, it can be seen that mechanical properties of polypropylene are much more susceptible to ionizing radiation and thermal stresses than the electrical properties are. The tensile properties should, therefore, be of significant interest in electrical insulation. Based on the changes in the mechanical and electrical properties, it can be assumed that the failure mechanism of the electrical insulator under multistress aging conditions could be a mechanical failure of the material, rather than direct homogeneous decay in the dielectric strength or thermal breakdown of the polymer.

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APPENDIX

**Effects of Multistress Aging (Radiation,
Thermal, Electrical) on Polypropylene**

Submitted to the

IEEE Transactions on Nuclear Science

October 1990

**EFFECTS OF MULTISTRESS AGING
(RADIATION, THERMAL, ELECTRICAL)
ON POLYPROPYLENE**

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ABSTRACT

Capacitor-grade polypropylene films were aged under multiple stresses (electrical, thermal and radiation) at the University at Buffalo 2 MW Thermal Nuclear Reactor. The aging experiments were performed for singular as well as simultaneous, combined stresses. The polypropylene was exposed to: combined neutron-gamma radiation with a total dose of 1.6×10^6 rad; electrical stress at 40V_{rms}/μm; and thermal stress at 90° C . Post aging diagnostics consisting of electrical, mechanical, physical and chemical characterization were carried out to identify degradation mechanisms for polypropylene film under multifactor stress aging.

The most pronounced changes were observed in the mechanical properties of the film. Significant decrease in elongation at break and tensile strength proved deterioration of the polypropylene under combined neutron-gamma radiation. This decrease was caused by chain-scission of the polypropylene molecules. It was found that the chain-scission was a process occurring at a greater rate than crosslinking due to the presence of oxygen in the radiation environment. The temperature stress had an opposite effect causing an increase in the above mentioned properties and offsetting, therefore, the negative effect of radiation. For the electrical properties, although changes were observed, they were not as significant as those for the mechanical characteristics. The breakdown voltage increased slightly because of radiation, while thermal and electrical stresses lowered it. A similar trend was also observed for volume resistivity. The combination of either thermal or electrical stress with radiation was overshadowed by the latter and overall increase in the above properties was observed. An increase in the degree of crystallinity of polypropylene was responsible for a slight improvement in the ac/dc electric strength as well as the volume resistivity. The effect of electrical stress was very little on both the mechanical and electrical

properties, due to relatively low applied field.

Based on this work it is found that the mechanical properties of polypropylene undergo much greater changes upon exposure to ionizing radiation and thermal stress than electrical properties. It can be concluded, therefore, that the failure mechanism of the electrical insulation under multistress aging conditions could be a mechanical failure of the material, rather than direct homogeneous decay in the dielectric strength or thermal breakdown of the polymer.

INTRODUCTION

As technology advances, increasing demands are made on materials and components. The most important requirement is the reliable operation under various operating and environmental conditions. Radiation has been added to the growing list of combined environments which already include temperature, electric stress, pressure, atmosphere, humidity, acceleration, shock and vibration.[1,2] There is need, therefore, for materials and components to perform reliably under this multistress environment.

Nuclear sources for electric power generation are well established for earth-based applications. Nuclear power sources are also promising candidates for future space power. As electrical power requirements increase with each succeeding space program, new technologies and often new energy sources must be used to provide the optimum power system. For long durations applications for which extremely high power levels (hundreds of kilowatts) are required, nuclear dynamic system is the most potential candidate.[3] With the numerous opportunities that the nuclear power generation offers, there is also a unique problem of self-generated neutron and gamma radiation. Also due to much higher power and energy densities of these reactors, and problems with waste heat disposal, thermal management also becomes a critical issue. The utilization of nuclear power as a source of energy for earth and space-based power sources provides, therefore, the designer with a radically different and challenging environment, one that includes a combination of high radiation, thermal and electrical stresses.

Organic products are listed among other materials needed for service in equipment located in areas where the radiation dose can be appreciable over the system design life. Presently, there are significant numbers of electrical components and seals in nuclear reactor environments requiring polymeric materials in their design and construction. A few of these components include power cables, capacitors, transformers, control cables, instrumentation cables, pump and fan driving motors, valves, relays and connectors.[4,5,6] Whatever the application, polymers and other organic materials are typically

found to be amongst the weak links in equipment where radiation aging is a factor. This is especially true in the case of an oxygen-containing atmosphere, where oxidative degradation can be induced by a number of different stresses including radiation, thermal, electrical, photochemical and mechanical stresses.[7] Thermally-induced aging is the most frequently encountered and studied aging stress. Predictions are considerably more difficult in environments where stresses other than thermal are dominant because aging methodology involving other stresses is less developed. In environments where two or more stresses are important, the situation is even more difficult.[7]

Considering the importance of multifactor stress on electrical insulation, the objective of this work is to find how a combination of radiation, electrical and thermal stresses affect the organic insulating materials. Polypropylene, being the primary organic dielectric used for capacitors located inside the containment, was chosen as the subject of this study. With a significant lack of information on the changes taking place in polymer insulation under influence of multistress aging (including electrical, thermal and radiation), this work sheds some light into this presently limited-data area.

EXPERIMENTAL ARRANGEMENTS AND PROCEDURES.

A special high voltage capacitor was designed in order to allow simultaneous exposure of polypropylene film to nuclear radiation, electrical and thermal stresses. A schematic representation of the test set-up showing combination of all three stresses applied to the capacitor is shown in Figure 1. The detailed description of capacitor design and individual stress conditions are given below.

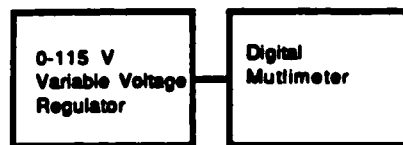
High Voltage Capacitor Design

High voltage capacitors were designed and wound using 25.4 μ m thick polypropylene film. Polypropylene was chosen because of its extensive use in high energy density capacitors. The capacitors for this work were manufactured to designed specifications by Maxwell Laboratories. A high voltage polypropylene wound capacitor is shown in its final form in Figure 2. Three major requirements had to be met in order to attain the goal of simultaneous stressing of polymer insulation:

- access to electrical stress application
- easy way of heat transfer to the volume of the capacitor
- restrictions on physical dimensions for radiation purposes

The capacitor design is shown in Figure 3. It had a PVC core with 2.7cm outer diameter and 7.6cm length. A structure consisting of two 25.4 μ m thick polypropylene films separated by two offset 9.0 μ m thick aluminum foils was wound on the core. The lengths of the films and foils were around 2.54m, giving

ELECTRICAL STRESS CONTROL



THERMAL STRESS CONTROL

TERMINAL BLOCK 1 (TB1)

5.5 m reusable leads
inside the standpipe

TERMINAL BLOCK 2 (TB2)

2.1 m disposable leads

RADIATION STRESS

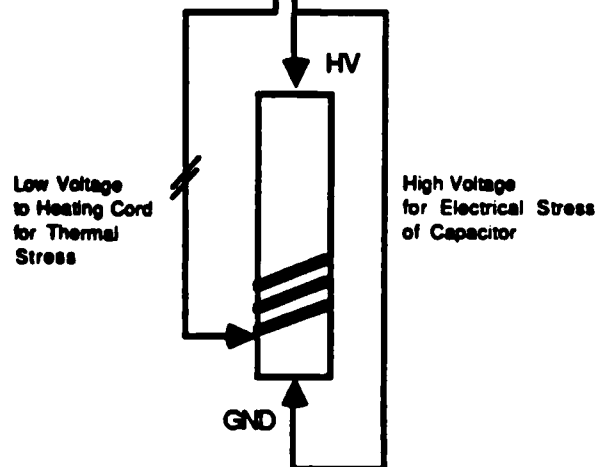


Figure 1. Experimental set up for multistress aging (E,T,R).

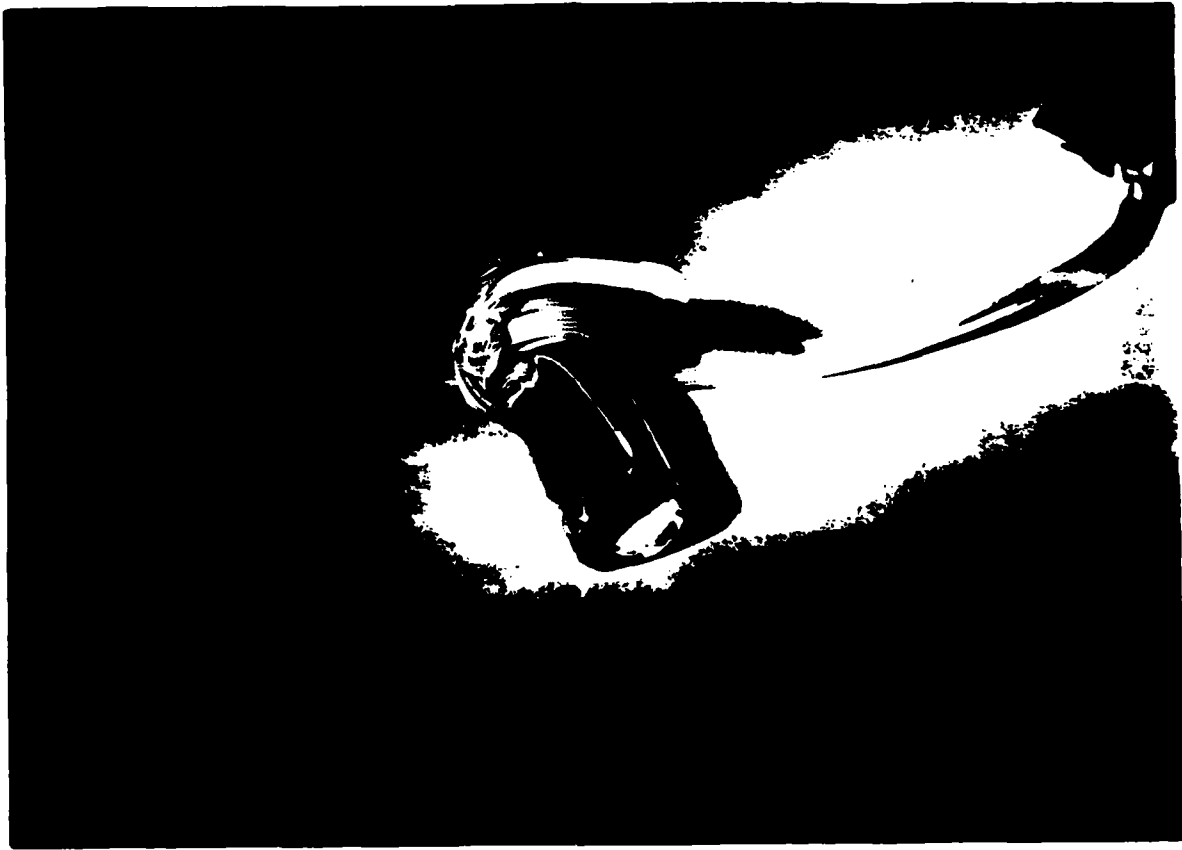
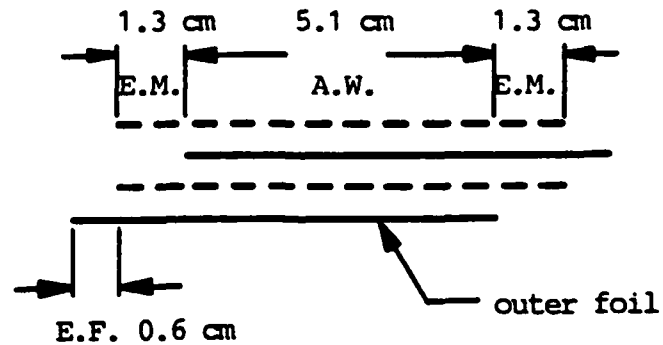
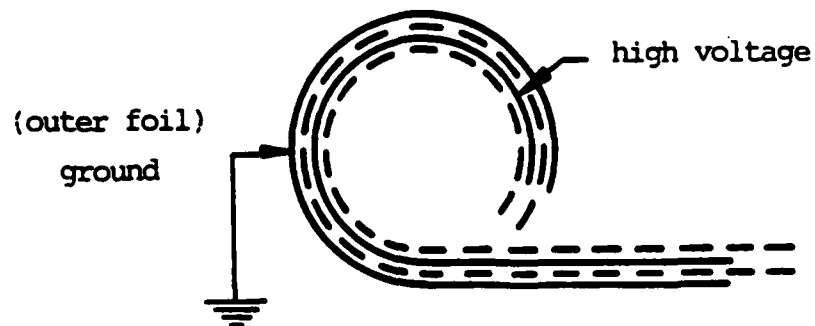


Figure 2. A high voltage polypropylene-wound capacitor used in the present experiments.

——— Aluminum foil ($9\text{ }\mu\text{m} \times 7.0\text{ cm}$)
 - - - - Polypropylene film ($25.4\text{ }\mu\text{m} \times 7.6\text{ cm}$)



(a) winding crosssection



(b) side view

Figure 3. High voltage polypropylene film capacitor design. (A.W. = Active Width,
 E.F. = Extended Foil, E.M. = End Margin)

the capacitor 28 winding turns. With a 5.1cm active width (AW=5.1cm), an average capacitance of 170nF was achieved. The winding was performed in such a way that the foil marked as outer foil in the Figure 3(b) became the outermost layer of the capacitor structure. The benefits of this design are as follows:

- the extended foils (EF=0.6cm) allow for easy application of electrical stress
- the outermost layer being aluminum allows for good heat transfer to the bulk of the capacitor and uniform temperature distribution
- finally, the outside diameter of the capacitor, being only 3.1cm, satisfied easily the physical constraints for radiation purposes

Test Capsule Design

In order to prevent any electrical short-circuit during irradiation inside the aluminum standpipe, the capacitor sample was fixed vertically on a Teflon rod inside the polypropylene container. The test capsule was attached with strings to Terminal Block 2 (TB2) as is shown in Figure 1. In addition, two pairs of leads 2.1 m long were attached to TB2 for both electrical and thermal stress of the sample. For high voltage connections, 20 gauge polyethylene insulated high voltage leads (60kVdc) were used. The voltage to the heating cord was supplied via a two-conductor UL listed cord. These 2.1m long leads plus the container and the heating cord were disposed after each radiation stress due to excessive residual radioactivity. On the other hand, the 5.5m long leads extending between TB1 and TB2, having negligible level of radioactivity, were reused in all radiation experiments.

The irradiated capacitors, after allowing sufficient time to cool down radioactively (2 months), were unwound and polypropylene films, subject to further testing, were stored in separate containers.

Nuclear radiation environment

A two megawatt, open-tank reactor at the Buffalo Materials Research Center was used for irradiation of the samples. From many different radiation facilities available at this site, the vertical standpipe was chosen for this study. The standpipe replaces the standard fuel elements in the fuel configuration and thus provides a dry chamber in the core accessible from the top. The tube extends from grid plate to the surface of the pool, equaling approximately to 7.6m. Using a standpipe as a medium of radiation provided two main advantages:

- high fluxes - 2×10^{10} n/cm²s - thermal neutrons
 1×10^9 n/cm²s - fast neutrons ($E > 3\text{MeV}$)
 1.5×10^6 rad/h - gamma radiation

- experimental flexibility, since experiments could be inserted or withdrawn easily and electrical leads were not obstructed

The standpipe had an inner diameter of 10cm, which was the major physical constraint on the samples being exposed. To achieve the maximum radiation effect on the stressed samples, the neutron flux within the standpipe was calibrated versus the distance from the bottom of the tube. The maximum radiation level inside the standpipe was thus found and the exact neutron flux versus neutron energy characterized at this point by means of Cobalt, Nickel, Magnesium and Aluminum Activation foils. The corresponding data is presented in Figure 4. Based on the measured energy spectrum of the neutron radiation the energy absorbed by the polypropylene film was calculated using procedure described elsewhere. [8] The energy absorbed by the polymer film due to the neutron radiation, however, accounts only for 7% of the total absorbed energy of 1.6×10^6 rad (during one hour radiation stress). Gamma radiation is a dominating factor as far as the energy absorbed by polypropylene is concerned.

Thermal Stress

The thermal stress of a capacitor sample was achieved by means of a small diameter flexible electric heating cord wrapped around the bottom part of the capacitor (in order to allow for unobstructed penetration of the ionizing radiation through the main part of the capacitor body). This is also shown in Figure 2. The aluminum foil being the outermost layer of the capacitor allowed for good heat transfer to the bulk of the capacitor. A woven fiberglass outer sheath of the heating cord in conjunction with the outside aluminum layer of the capacitor being at ground potential, prevented any electrical hazards during the electrical stress.

For thermal stressing of the capacitor specimen, the temperature was selected to 90°C, which corresponds with the maximum upper service temperature for polypropylene.[9] The temperature inside the capacitor structure was measured by means of small diameter flexible temperature probe and Cole-Palmer digital thermometer. The initial calibration of the temperature within the windings was performed in the laboratory at 43°C (ambient temperature inside the standpipe) using NAPCO thermostat regulated oven.

Electrical Stress

To electrically stress the capacitors, a 60Hz ac voltage with the magnitude of 1kVrms was applied to the opposite extended foil edges of the windings. This corresponds to a stress of 56V/μm, which is the maximum operating voltage stress for polypropylene in the present state-of-the-art capacitors. [10]

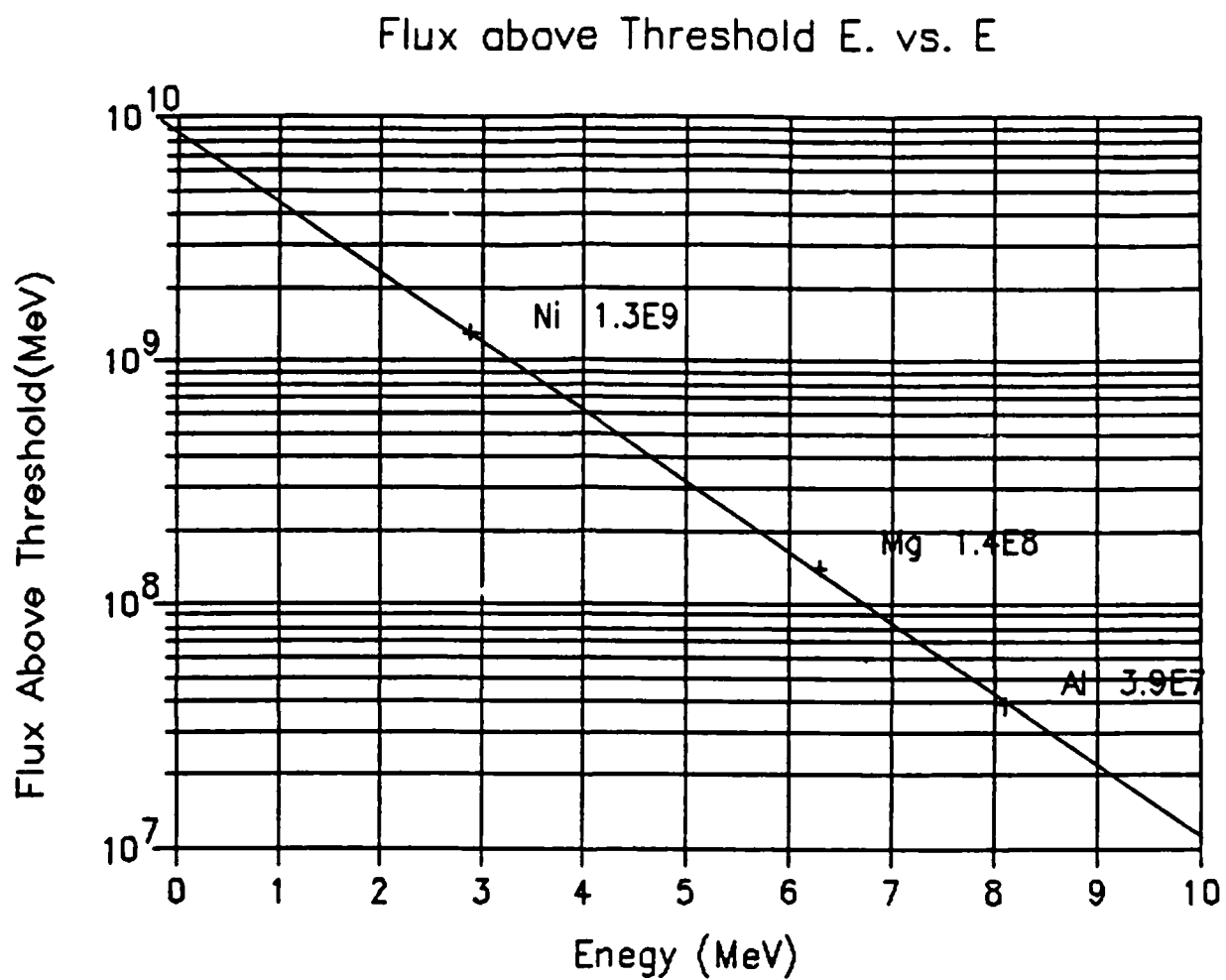


Figure 4. Neutron flux above threshold energy versus energy. (Plotted for 10 cm standpipe)

The electrical stress of the capacitor at the remote site of nuclear reactor was achieved with a portable step-up transformer (5kV, 300mA rating). During the electrical stress, the voltage on the secondary winding of the transformer was monitored with the 2213A Tektronix scope and Keithley digital multimeter, after 1000:1 reduction with P6015A Tektronix high voltage probe.

THE EFFECTS OF MULTISTRESS AGING ON ELECTRICAL PROPERTIES OF POLYPROPYLENE

The experimental results reflecting the effects of singular and combined stresses (radiation, thermal, electrical) on electrical properties of polypropylene film are shown in Table 1. From both ac and dc breakdown data, it can be seen that both thermal and electrical stresses, in general, tend to lower the breakdown voltage. On the other hand, a slight increase in the dc and ac breakdown strength of polypropylene was observed for irradiated samples. This increase can be attributed to a rise in the crystalline fraction of the polymer as compared to the amorphous portion. This observation was confirmed with two diagnostic techniques, namely differential scanning calorimetry and wide angle X-ray spectroscopy, results of which are presented later. Based on the dc breakdown voltage data for polypropylene subjected to combined multiple stresses of radiation (R), temperature (T) and electric (E) field, it is observed that these stresses have an interactive (synergistic) effect. The dc breakdown voltage is much lower for combined E/T/R stresses than expected by merely adding the effects of each stress accounted individually. Apparent synergistic effects were also reported earlier for combined environments of radiation and elevated temperature by Clough and Gillen.[11,12]

The dielectric characterization of aged polypropylene film consisted of: dielectric constant, dissipation factor and volume resistivity. The post-stress characterization of the polypropylene film samples showed no distinguishable change in both the dielectric constant and the dielectric loss. It is believed that the two month period of radioactive cooling down of the samples eliminated any temporary effects and allowed for recovery of the dielectric properties of the polypropylene. The variations in the volume resistivity of polypropylene films, on the other hand, were more pronounced (Table 1). Both thermal and electrical aging resulted in a decrease of volume resistivity, -50% and -30%, respectively. In contrast, radiation stress caused a 17% increase of this dielectric property. Similarly to the improvement of ac and dc breakdown strength of polypropylene subject to radiation aging, increase of the volume resistivity is attributed to the increase in the degree of crystallinity.

TABLE 1
ELECTRICAL PROPERTIES *

Radiation Stress 1.6x10⁶rad	Thermal Stress 90°C	Electrical Stress 40Vrms/μm	AC Breakdown Voltage (kV)	DC Breakdown Voltage (kV)	Volume Resistivity (Ωcm)
-	-	-	9.05 (.23)	15.71 (1.81)	11.2 (2.1)
-	X	-	8.59 (.47)	15.22 (1.77)	5.61 (1.2)
-	-	X	9.09 (.19)	15.26 (2.52)	7.81 (1.4)
-	X	X	8.61 (.37)	15.38 (1.96)	7.58 (2.11)
X	-	-	9.12 (.38)	16.67 (1.94)	13.1 (3.2)
X	-	X	9.55 (.68)	16.79 (1.30)	11.9 (2.4)
X	X	-	9.43 (.49)	15.36 (2.41)	13.9 (2.0)
X	X	X	9.27 (.34)	14.15 (1.73)	10.8 (2.4)

* The value in parenthesis represents standard deviation.

THE EFFECTS OF MULTISTRESS AGING ON MECHANICAL PROPERTIES OF POLYPROPYLENE

The results of post-stress mechanical characterization of polypropylene film are grouped in Table 2. In contrast to the electrical properties, the changes in mechanical properties are much more pronounced. A 13% decrease in the ultimate tensile strength and 25% decrease of elongation-at-break $\Delta l / l$ of polypropylene is observed after subjecting the film to nuclear radiation stress for a period of one hour. Thermal stress, on the other hand, resulted in a slight increase of these mechanical properties. It is very interesting to note that radiation and thermal stresses, having opposite effects on the ultimate tensile strength and $\Delta l / l$ neutralize each other to some extent when both stresses are applied simultaneously. As for the Young's modulus, singular stresses alone had very little effect on this tensile property. However, the synergistic effect in the degradation of polymer under combined stresses is very evident. The following combination of stresses: E/T, T/R, E/T/R resulted in significant -5%, -10% and -11% decreases of Young's modulus, respectively.

All of the above tensile properties, which are of utmost importance in the mechanical aspect of dielectric film's application, are very susceptible to ionizing radiation and to thermal stress. When the polypropylene film is subject to multifactor stress aging, the dielectric properties such as breakdown strength, change insignificantly (<6%), while the mechanical properties change drastically (up to 25%). The mechanical properties, therefore, should be of significant interest as far as electrical insulation is concerned.

PHYSICAL AND CHEMICAL CHARACTERIZATION OF POLYPROPYLENE SUBJECT TO MULTIFACTOR STRESS AGING.

CRYSTALLINITY

Two techniques were utilized in determination of the crystallinity of the polypropylene film, namely wide-angle X-ray diffractometry and differential scanning calorimetry (DSC). For the X-ray diffraction patterns, the intensities of the beams diffracted by crystalline regions were isolated from that of amorphous regions using the procedure described elsewhere[13,14]. The areas of crystalline (A_c) and amorphous diffractions (A_a) were used to calculate the percent crystallinities:

$$W_c = [A_c / (A_c + K A_a)] 100\% \quad (1)$$

TABLE 2
MECHANICAL PROPERTIES *

Radiation Stress 1.6x10 ⁶ rad	Thermal Stress 90°C	Electrical Stress 40V _{rms} /μm	Ultimate Tensile Strength (x10 ⁸ N/m ²)	Elongation-at-Break (%)	Young's Modulus (x10 ⁹ N/m ²)
-	-	-	2.131 (.063)	86.2 (4.3)	2.68 (.23)
-	X	-	2.268 (.075)	94.5 (4.3)	2.74 (.28)
-	-	X	2.152 (.086)	87.8 (7.5)	2.59 (.12)
-	X	X	2.184 (.086)	88.7 (3.7)	2.53 (.18)
X	-	-	1.732 (.034)	65.0 (6.0)	2.61 (.06)
X	-	X	1.812 (.057)	67.8 (2.4)	2.61 (.17)
X	X	-	2.00 (.031)	84.8 (3.7)	2.40 (.13)
X	X	X	1.888 (.067)	87.5 (2.6)	2.37 (.20)

* The value in parenthesis represents standard deviation.

Differential scanning calorimetry allowed to determine the melting point, the heat of fusion of polypropylene samples, and the percent crystallinity.

$$W_c\% = (\Delta H_f / \Delta H_{fc}) \cdot 100\% \quad (2)$$

Where, ΔH_f is the heat of fusion of the sample and

ΔH_{fc} is the heat of fusion of a 100% crystalline polypropylene

($\Delta H_{fc} = 209 \text{ J/g}$ for isotactic polypropylene)

Table 3 compares weight percent crystallinities using both techniques. The DSC measurements show on the average 6% lower readings than the crystallinity values obtained from X-ray technique. Such discrepancies are not unusual and differences up to 17% were observed in earlier studies, especially at higher crystallinities.[15] For both techniques, however, it can be seen that the crystallinity of the polypropylene film subjected to nuclear radiation increases slightly. When combined with other stresses such as thermal and electrical, this increase is even more pronounced. A similar increase in the degree of crystallinity under small radiation doses in open air has been reported for polyethylene and polypropylene.[16,17] No specific trend has been observed in the melting point of polypropylene film as a function of multistress aging.

An increase in the crystallinity of polypropylene under combined stresses involving radiation is believed to be responsible for the slight improvement in the ac/dc dielectric strength and volume resistivity, as was reported earlier.

CHAIN SCISSION

The partial extraction of polypropylene films with boiling toluene [$T_b = 111^\circ\text{C}$] showed that this polymer undergoes chain scission when subjected to radiation, thermal and electrical stresses in open air environment. A reduction in the gel fraction of extracted polymer is evident, although not very significant. The long chains of polymer molecules in the presence of molecular oxygen disproportionate and lead to chain scission. This process is taking place along with crosslinking; however, the results of gel fraction analysis show that the chain scission is a process occurring at a greater rate than crosslinking. Chain scission of polypropylene molecules is responsible for degradation in the mechanical properties of polypropylene, such as ultimate tensile strength and elongation at break.

TABLE 3
PHYSICAL PROPERTIES

RADIATION STRESS 1.6X10 ⁶	THERMAL STRESS 90°C	ELECTRICAL STRESS 40Vrms/μm	DIFFERENTIAL SCANNING CALORIMETRY			X-RAY Crystallinity %
			Melting Point °C	ΔHf	Crystallinity %	
-	-	-	161.6	87.3	41.7	47.3
-	X	-	168.1	85.3	40.8	46.8
-	-	X	167.0	87.4	41.8	47.1
-	X	X	165.5	85.7	41.0	46.7
X	-	-	163.9	93.9	44.9	49.0
X	-	X	165.7	87.4	41.8	50.4
X	X	-	165.4	86.9	41.6	46.9
X	X	X	164.9	93.2	44.6	51.5

INFRARED SPECTROSCOPY

In order to find out if oxidation or any chemical changes were taking place, the polypropylene films subjected to multistress aging were analyzed using infrared spectroscopy (IR). The samples were scanned from 2.5 to 25 μ m (4000 cm^{-1} to 900 cm^{-1} wavenumber).

Two peaks are expected if oxidation was to take place, one around 1720 cm^{-1} corresponding to carbonyl group C = O, and another at 1102 cm^{-1} corresponding to C-C stretching vibration in the



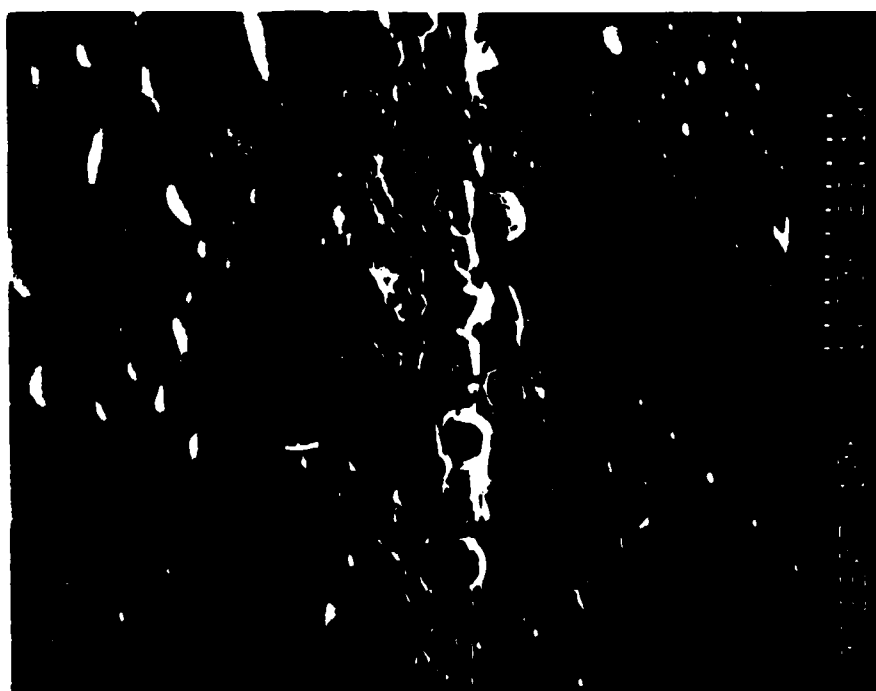
None of these two peaks were detected in the infrared spectra. The relatively short time and low dose of radiation stress resulted in non-detectable level of oxidation of the multistressed polypropylene, which would otherwise be expected.

SCANNING ELECTRON MICROSCOPY

The surface topography of the virgin, single- and multi-stressed polypropylene films are shown in Figure 5. The surfaces of the insulating films were specifically analyzed at the edges of aluminum foils, where the electric stresses are the highest. The high electrical stress, due to nonuniformity of the electric field at the sharp edge of aluminum electrode, caused the biggest single damage to the polypropylene. The high voltage partial discharges taking place in the region of enhanced field stress created local hot spots and caused the formation of structure as shown in Figure 5(a) (Combined electrical and thermal stress). Similar observations were made earlier for polypropylene film subjected to high voltage partial discharges.[20] The damage was even more pronounced when the electrical stress was combined with radiation, shown in Figure 5(b), where physical punctures are visible at the foil edge. The analysis of the polypropylene film in the area of uniform electrical stress showed no change in the polypropylene surface topography.



(a)



(b)

Figure 5. SEM photographs of polypropylene film at the aluminum foil edge.
(a) combined electrical and thermal stress
(b) combined electrical and radiation stress

CONCLUSIONS

The effect of multifactor stress aging on electrical, mechanical and physico-chemical properties of capacitor grade polypropylene film was studied in order to characterize the nature of changes and to identify degradation and failure mechanisms. The electrical properties measured comprised of dc/ac breakdown strengths, volume resistivity, as well as in-situ and post-stress dielectric properties. Both thermal and electrical stresses decreased slightly the ac and dc breakdown voltages of polypropylene. The radiation stress, on the other hand, increased the breakdown strength of the polymer for both ac and dc electric field stresses. A similar trend was also observed for volume resistivity. The radiation stress is the most dominant factor influencing both the electric strength and the volume resistivity. The combination of either thermal or electrical stress with the radiation is overshadowed by the latter and overall increase in the above properties is observed. This increase is attributed to the increase in the degree of crystallinity of the polypropylene.

The most pronounced changes were observed, however, in the mechanical properties of the film. There is a significant decrease in elongation-at-break and tensile strength proving deterioration of the polypropylene under combined neutron-gamma radiation. This degradation is attributed to chain scission of the polypropylene molecules. The temperature stress had an opposite effect causing an increase in the above mentioned properties and offsetting, therefore, the negative effect of radiation. In addition to the discussed changes, strong interactive (synergistic) effects are observed. The dc breakdown strength and the Young's modulus are much lower for combined E/T/R stresses than expected by merely adding the effects of each stress individually.

Based on this work, it can be seen that mechanical properties of polypropylene are much more susceptible to ionizing radiation and thermal stresses than the electrical properties are. The tensile properties should, therefore, be of significant interest in electrical insulation. Based on the changes in the mechanical and electrical properties, it can be assumed that the failure mechanism of the electrical insulator under multistress aging conditions could be a mechanical failure of the material, rather than direct homogeneous decay in the dielectric strength or thermal breakdown of the polymer.

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Dear Prof. Laghari:

The recommendations of the Reviewers of your paper, "Effects of Multistress Aging (Radiation, Thermal, Electrical) on Polypropylene," have now been received.

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