NISTIR 4486

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INTRODUCTION

One of the current problems in the operation of nuclear power plants as they age is the estimation of the lifetime of the electrical insulation used inside the reactor confinement. This insulation, having been manufactured for installation in reactors, has been subject to lifetime testing prior to certification for use. This program has been quite successful, as there have been too few failures to make any changes in the requirements.

As the plants age, however, the question has arisen as to whether an estimate can be made as to the remaining life left in the cable insulation, so that the cables can be used beyond their original certified life. If this were possible, costly and difficult overhauls of the reactor wiring could be avoided. Given the conservative nature of the certification process, this possibility of extended lifetime certainly exists.

The original tests on the cables were acceptance tests, where the cables were subjected to an environment more severe than found in service and they had to maintain their integrity. They have lead to the situation where cables are used possibly for only a small part of their potential lifetime. For estimation of a lifetime, such tests provide little information other than a possible minimum allowable exposure. These tests do not follow changes in the cables as they age. The effects of accelerated aging, used in these tests, are not studied. Finally, there is no indication at all of the ultimate failure mechanisms of the cables and when and how they might occur.

Complicating lifetime estimation is the requirement that any electrical cable must remain functional during a LOCA (Loss of Coolant Accident). Since such an event can put a severe thermal and radiation stress on the cables inside a reactor, the cables must not be too close to failure prior to such an event. This means that a cable at the end of its useful life could appear to be quite normal since its mechanical and electrical properties may change quickly only when the insulation is near failure.

The combination of all these considerations makes lifetime estimation very difficult for reactor cables currently in use. Not only is a convenient measure of age difficult to establish, but, also, any measure must be obtained in a relatively harsh environment. Furthermore, any estimation must attempt to predict the future, including the possibility of an event more severe with respect to lifetime than the total environmental aging allowable to a real endpoint. This report will detail the work at NIST that has been undertaken to see if the method of Time-Domain Dielectric Spectroscopy can evaluate the aging in reactor cables with the goal to ultimately estimate lifetimes.

Nature of the Problem

The degradation of electrical insulation under combined thermal and radiation stresses is a well known phenomenon. Unfortunately, the rate at which this degradation occurs is highly material dependent and a small amount of additives can make a large difference. In addition, the mechanisms are very complex and highly varied. These factors make theoretical prediction of lifetimes very difficult. They also create the possibility that there can be relatively large variations in the useful lifetime in a given material.

If polymeric electrical insulation used in reactors is to be used beyond its originally certified lifetime, it clearly will be used that much closer to the total amount of time that they can be used safely.

Since different pieces of insulation will be subject to different aging conditions, a lifetime based on a worst case exposure may be overly strict. Also, variations in material response will become more important.

The approach undertaken in this investigation has been to see if some measure of cable aging can be found. Ideally, this measure should vary smoothly with aging. It should, if possible, be able to resolve the contributions from different sources of aging as they could interact or lead to different failure mechanisms. The measure should have clear changes that are above any possible sample to sample variations in order to prevent a difficult measurement problem from becoming impossible. Finally there must be some possibility for instrumentation without major modifications to any reactor.

Potential of Dielectric Measurements

Much of the chemistry of radiation damage involves the generation of charged fragments in an otherwise nonconducting environment. Chemical oxidation associated with exposure to high temperatures can also create similar results. While the chemistry involved in the two processes can be quite different, the presence of charged species will have a large effect on the dielectric properties of the insulation. If the species can migrate through the insulation, a DC conductivity will be observed. If they are bound, a dipolar type relaxation can occur. Any interactions with the conductors can lead to large Maxwell-Wagner type relaxations.

In addition to these changes, the mobilities themselves will be a function of the state of the insulation. Polymers are not hard, rigid solids and, as they age, mobilities can change quite drastically. Dipolar charge responsible for dielectric relaxation behavior tends to follow closely the time scales for mechanical relaxations. As a material becomes hard and brittle, these relaxations tend to evolve to longer times. If characteristic loss maxima can be identified, then such aging could be readily followed.

Just as important is the fact that the dielectric behavior of a material is measured by the response to a small applied field. This measurement is inherently nondestructive to the measured sample. Methods have been developed such that, at the low frequencies that are expected to be important in this problem, the length of the leads used to connect the sample to the measuring apparatus do not interfere with the measurement. Since electrical measurements form the basis of most modern instrumentation, they clearly offer the possibility of automation and remote sensing.

Time-domain Dielectric Spectroscopy

Most of the changes in the dielectric properties of polymeric insulation that are expected to be important to age monitoring are usually found in the low part of the frequency scale. This scale extends from kilohertz down to lower frequencies. Furthermore, in order to sort out changes resulting from temperature as well as different relaxation mechanisms, a wide range of frequency can be necessary. For conventional, fixed frequency devices, such as bridges, covering a large frequency span can be very time consuming. The problem becomes worse if very low frequencies are needed so that just a single cycle can take a long time.

The Time-Domain Dielectric Spectrometer is a method for measuring dielectric properties over the frequency range from 10^4 to 10^{-4} Hz by obtaining the sample response to a step application of voltage.

The data are then converted to the frequency domain by a numerical Laplace transform. This method allows acquisition of the entire frequency range, typically at least 7 decades, in a time that is less than that required for one cycle of the lowest frequency of interest. Since the measurements are capable of high precision and sensitivity, if different relaxation mechanisms are present, they can distinguish between them.

The clearest example of this resolution will be seen in the ability to separate relaxation processes from the contributions of ionic conduction in the polymeric insulation. Since polymers always have some loss at low frequencies and, since irradiation of polymers can lead to the generation of charge carriers in the bulk of the material, a measurement at a single frequency can be misleading. A frequency scan can lead to a separation of the two contributions and provide a level of information that would be missing from a single frequency measurement where the two contributions would simply add. This separation will be emphasized especially in what follows with respect to the interpretation of the data.

This report will detail the work done at NIST to establish the connection between cable aging in a reactor environment and the results observed by the Time-Domain Dielectric Spectrometer. The results will show that cable aging can be observed and followed. On the one set of cables where a systematic aging protocol was made, clear, readily distinguishable results were obtained. Despite not having a good end of life criterion available, it will be seen that dielectric loss measurements of the cable insulation have the ability to monitor cable aging. While the endpoint question will not be addressed in this report, these results suggest that if such a criterion can be agreed on, dielectric measurements could be used to establish a lifetime estimation.

RADIATION EFFECTS

In order to interpret dielectric measurements in polymeric insulation, some knowledge of the process of aging is necessary. This knowledge can help in the interpretation of the measurements as well as act as a guide as to what results could be expected. For dielectric measurements in particular, the general nature of radiation damage leads to the expectation that there should be a definite evolution in both loss and conductance with radiation. However, as indicated in the following sections, the relationship need not be simple and is quite material dependent.

Degradation

When ionizing radiation with energies above 100 eV passes through a polymer or any other material, its energy is absorbed and the absorption can cause a molecule to enter highly excited states leading to severe chemical change. There is more than enough energy to cause bond rupture and degradation. Which reaction products will be formed are a function of the exact nature of the species involved.

The complexity of the interaction of radiation with organic materials is well illustrated in the monograph by Földiák on the hydrocarbons[1]. The chemistry is very involved and dependent upon the exact chemical nature of the compound. For aliphatic, saturated compounds, there is a great deal of hydrogen liberation and bond rupture. Unsaturated hydrocarbons when irradiated have less bond rupture, but instead react to form higher molecular weight species. Aromatic compounds such as benzene and toluene tend to have much greater stability towards radiation damage. In all cases, there is ionization and subsequent reaction to form other ions, free radicals and excited states.

These excitation products are very reactive and unstable. They often lead to degradation and smaller molecules[2]. If oxygen is present, these excited species will readily react to form carbonyls, alcohols and acids. These latter species all can contribute to a measured dielectric response through ion formation and the presence of large dipoles. If chlorine containing compounds are present, the chlorine will preferentially split off with the possibility of new polar or ionic species being formed[2]. If the fragments are small enough, they could lead to loss of mass and shrinkage of the original material.

Cross-linking

The radiation induced reactions just mentioned lead to very reactive species at least for a short period of time. In a solid material, these species can then interact with neighboring molecules. In the case of polymers, these reactions can lead to cross-linking. These cross-links, if they proceed far enough, will create large changes in the behavior of the polymer. A flexible polymer will become rigid and brittle. Softening points will shift to higher temperatures. Polymeric relaxation processes will drastically shift their time scales.

For a given polymer, whether chain scission and subsequent degradation or whether cross-linking occurs is a function both the polymer and the conditions under which it is irradiated[2,3]. Polyethylene undergoes cross-linking while polyvinyl chloride can either degrade or cross-link or have both processes occur at the same time. There can even be a reactive bonding to adjacent surfaces[2]. If fillers are present, this bonding can alter the physical properties of the polymer. In some of the electrical cables examined in this report, there was clear bonding between originally separate layers.

Secondary Reactions

The immediate products of irradiation include free radicals and ionic species that are quite reactive. They can be detected by many techniques and do not necessarily react immediately. Free radical decay in polyethylene has been studied quite intensively[4]. The decay rate is a function of the exact nature of the polyethylene as well as the temperature. Even stress can affect the decay rate. This has been specifically noted in polyethylene[5]. Similarly, the ionic species can be followed by the decay of the radiation induced conductivity[6].

These reactions can lead to complicated molecular structures that would not be expected from the starting material. This is clearly evidenced in polyethylene, where irradiation induces a yellow color[2]. It also leads to the consequence that the aging of a polymer can be quite sensitive to small amounts of an added material. Traditionally, this would include antioxidants and other processing agents, but it could include anything other than the base resin. In a series of tensile tests on a colored polyethylene cable of the type used prior to the mid-1970's, a pronounced correlation of the rate of degradation with color was found[7].

These secondary reactions can also affect the tendency for a polymer to degrade or cross-link. For ethylene-propylene rubber (EPR), the presence of agents such as tetravinyl silane, chlorobenzene, nitrous oxide, allyl acrylate and neopentyl chloride significantly enhanced the polymer's tendency to cross-link[8]. Polyvinyl chloride normally degrades with radiation by dehydrochlorination with intense color formation. The addition of multifunctional groups can change that into cross-linking[9]. In the presence of oxygen, oxidation with large modifications to its behavior can occur[10]. The presence of flame retardants has been found to reduce the rate of cross-linking in low density polyethylene[11].

These variations with the exact state of the polymer show that a given polymer could age significantly differently under different conditions. There is also a possibility of batch to batch variation. These considerations suggest that a sensitive monitor of aging is desirable. Since most of the reactions involve species that respond electrically, and since dielectric properties tend to follow mechanical relaxation behavior, a dielectric test would seem to offer a good hope for evaluating remaining lifetime. The importance of such studies has been stated previously[12].

DIELECTRIC MEASUREMENTS

Fundamentals

In order to properly discuss the results obtained at NIST using the Time-Domain Dielectric Spectrometer, some of the fundamentals of what is measured must be reviewed. This includes the determination of dielectric constant and loss with its relation to measurable quantities, as well as the relation of dielectric constant and loss to the structure of the material. These considerations become more important than with ordinary measurements since the instrument bridges the usually distinct domains of time and frequency.

The basis for any dielectric measurement is the measurement of capacitance, since the fundamental definition of a dielectric constant, ϵ, is the ratio of a capacitance between a pair of conductors with the measured substance completely filling space around them, C, and with vacuum, C_g. The common definition is for static fields with the capacitance C, defined as

$$C = Q/V , \tag{1}$$

where Q is the stored charge and V is the applied voltage. For a sinusoidal applied voltage of frequency ω, the definition becomes

$$C(\omega) = I/\omega V , \tag{2}$$

where I is the measured current. Since a sinusoidal voltage is characterized by both amplitude and phase, the definition of ϵ, modified to include both, is the complex dielectric constant ϵ^*,

$$\epsilon^* = \epsilon' - i\,\epsilon'' = C(\omega)/C_g . \tag{3}$$

The term ϵ'' is called the loss index or dielectric loss as it represents the only contribution to the dissipation of energy.

Normally, capacitance measurements are not made through the measurement of current and voltage. Instead, they are often made with some form of admittance bridge. Such a bridge represents the measurement as the equivalent combination of a parallel capacitance and conductance, $C_m + G_m$. This provides the equivalent definition of ϵ^* as

$$\epsilon^* = (C_m - i\,G_m/\omega)/C_g . \tag{4}$$

An important point to realize is that any admittance, at a given frequency, can be represented as a parallel combination of a conductance with a capacitor and inductor. Therefore, for a material with dielectric constant ϵ^*, if there are processes that can be associated with both a dipolar loss mechanism as well as with ionic conductivity, a single frequency measurement can only determine the sum of the two. Any separation would have to be done as a result of multiple frequency measurements and interpretation.

For use in monitoring cable insulation, ϵ^* must be related to the properties of the insulation. In terms of the molecular properties of the insulation, this connection has been well established [13]. The magnitude of ϵ is a function of the number and strength of any permanent dipoles, μ, present in the material. For homogeneous materials, the Fröhlich-Kirkwood-Onsager equation,

$$\epsilon_s - n^2 = \frac{3\epsilon_s}{2\epsilon_s + n^2} \left[\frac{n^2 + 2}{3}\right] \frac{4\pi N_0 \mu^2}{3kTV} g \,, \tag{5}$$

where N_0 is Avogadro's Number, k is Boltzman's constant, V is the molar volume, T is the absolute temperature, ϵ_s, the low frequency limiting value of ϵ', is the equilibrium dielectric constant, n is the refractive index, and g is the average neighbor to neighbor correlation factor between dipoles. For a solid in which the dipoles can only rotate over a limited range of spatial angles, μ is replaced by its maximal projection parallel to the applied field.

At frequencies high enough so that the dipoles start to lose their ability to follow the electric field, ϵ' starts to decrease and dielectric loss, ϵ'', starts to appear. Ultimately, as frequencies increase, the loss passes through a maximum and ϵ' reaches a new limiting value, ϵ_∞, corresponding to n^2 in Eq. 5. The frequency of maximum loss, ω_{max}, is related to the reciprocal of the time it takes for the dipoles to reorient in a changed field, the relaxation time τ. The magnitude of the dispersion is equal to the difference between ϵ_s calculated for full orientation and ϵ_∞ calculated for any motion remaining on time scales short compared to the mode of relaxation being considered. If there is a conduction process present, the conductivity, σ, has an additive contribution to ϵ^* of the form $- i\, \sigma/\epsilon_0\omega$, where ϵ_0 is the permittivity of free space. This contribution is proportional to both the ionic concentration and the ionic mobility. The mobility, in turn depends on both the ion and the nature of the medium.

Relation to Aging

The ability of any dielectric technique to follow aging is dependent on the sensitivity of its parameters to the aging process. From the brief discussion just given, changes in both the chemistry and exact physical state of the system can be expected to influence these properties. The utility of dielectric techniques to monitor lifetime will depend not on the ability to see changes as much as these changes being clearly resolvable, being capable of being measured and being tied to a clear criterion for useful life.

Physical aging

It has become customary to consider any irreversible changes in the physical state of a system that are not chemically induced, physical aging. For an amorphous polymer below its glass transition temperature, there is a gradual evolution to a more dense state at fixed temperature and pressure as any trapped excess volume relaxes. Typically this relaxation occurs on time scales long compared to other time scales such as time to creep or the time for a dipolar relaxation process. The consequence of this aging process is for observable relaxations to gradually move to longer time scales. For dielectric relaxations, as well as mechanical relaxations, this means that there will be shifts in the frequencies of maximum loss to lower values. These changes are observed in most polymeric systems.

There can be aspects of physical aging other than time scale shifts. If the polymer is semicrystalline or can crystallize, then changes in the degree of crystallinity or even the microstructure can occur. These changes can lead to quite complex behavior as a function of time. While the general trend is to slower relaxation processes, there now arises the possibility for changes in the strength of the relaxation or even the creation or disappearance of relaxations.

A final consequence ot physical aging is that, since the exact physical state of the material affects any relaxation process, a change in the ambient conditions will also affect what is observed for a relaxation. This includes both temperature and pressure. The typical changes in atmospheric pressure are too small to be observable, but the changes with temperature are very large. The usual equation governing this change, the Arrhenius equation, is given by

$$\omega_{max} = A \exp(-\Delta H_a/RT) , \qquad (6)$$

where ΔH_a is the activation energy associated with the loss process and R is the ideal gas constant. For relaxations related to a glass temperature, the Arrhenius equation is not followed but rather the Vogel or WLF equation,

$$\omega_{max} = B \exp(-\Delta H_a/[R(T - T_0) , \qquad (7)$$

is more accurate, where T_0 is a temperature a little below the glass temperature. If conductivity is being followed instead of a loss maximum, the same governing equations must be used except the conductivity, σ, replaces ω_{max}. As a final comment, ΔH_a, itself, can change upon aging.

Chemical aging

The effects arising from physical aging can be amplified considerably by chemical changes upon aging. For a polymeric material there can be oxidative degradation, thermal decomposition, hydrolysis, and loss of plasticizer or other additive. Not only can degradation occur but also cross-linking. The changes can be similar to those found in physical aging, but are often larger and more varied. There can be large changes in tensile strength and elongation to break. The material can become quite brittle or even, in certain circumstances, become quite gummy.

What is important for condition monitoring is that in chemical aging the changes are being forced by a chemical change. This can lead to not only changes in relaxation times but also amplitudes as the number and strength of the dipoles present change. The same is true for conductivity, as not only is the mobility of the ions being changed, but also possibly the number. Therefore chemically induced changes should be readily observable dielectrically.

Radiation effects

The effects of radiation on aging are expected to be similar to those of chemical aging. The main difference is, as outlined above, that the species generated by radiation are more reactive than those generated by being exposed to ordinary ambient conditions. There are many ionic species capable of being generated. Decomposition can be accelerated with the generation of small fragments. The presence of free radicals can lead to unusual molecular rearrangements and extensive cross-linking. Many of these reactions would not take place in the absence of radiation.

It is the difference in reaction products that allows the hope that dielectric measurements could distinguish between thermal and radiation damage. There is the possibility of observable differences in relaxation behavior as well as differences between the relative changes in relaxations and conductivity. The ability to make this separation is dependent not only on measurement capability, but also on the material behavior itself. For PVC, where there are only small differences in aging[9], there would not be any expectation of being able to find much in the way of a difference.

Frequency Spectrum

The response at any given frequency is the sum of all relaxation processes present as well as a possible conduction term. The only way there can be a possibility of separating the different contributions to the response is if there is sufficiently broad frequency coverage so that the differing frequency responses can be separated. This is a technique that is common with most ordinary spectroscopic techniques.

What makes the problem difficult is the observation that the sharpest response possible is the Debye relaxation function

$$\frac{\epsilon^{*}-\epsilon_{\infty}}{\epsilon_{s}-\epsilon_{\infty}}=\frac{1}{1+i\,\omega\tau}=\frac{1}{1+\omega^{2}\tau^{2}}-i\,\frac{\omega\tau}{1+\omega^{2}\tau^{2}}\,, \tag{8}$$

which has a width at half maximum for ϵ'' of over a full decade of frequency. Polymeric materials have relaxations that are even broader and take a larger frequency band to cover. If an observable conductivity is present, then in order to determine its value there must be enough frequency coverage to establish a $1/\omega$ behavior.

Especially where the expected behavior is not known, a single frequency measurement may yield paradoxical results where loss levels can go through reversals as a function of time even though the underlying behavior is continually evolving. Single frequency measurements may be able to be employed if the expected behavior is well known, but that must first be established.

Ordinary constant frequency techniques have trouble with the large frequency range that is desirable to establish dielectric relaxation. Bridges ordinarily cover at most 3 decades at audio frequencies. Each point takes an appreciable length of time for balance so that even these decades take a long time to measure. Many features, especially ionic conductivity become apparent at even lower frequencies, where it can take a long time to wait just for a single cycle. These considerations have lead to the development of the Time-Domain Dielectric Spectrometer and the investigation of it as a condition monitoring tool for cable insulation.

Time-Domain Dielectric Spectrometer

The Time-Domain Dielectric Spectrometer[14,15] has been developed to maximize the frequency band for dielectric measurements while preserving measurement accuracy. Instead of measuring the dielectric response point by point, it measures the charge response of a sample to a step voltage as function of time as shown in Figure1. If the charge Q(t) is determined for the applied voltage E^0, then one can define a time dependent capacitance $C(t) = Q(t)/E^0$. Then the complex frequency-dependent capacitance $C^* = C' - i\,C''$ is given by

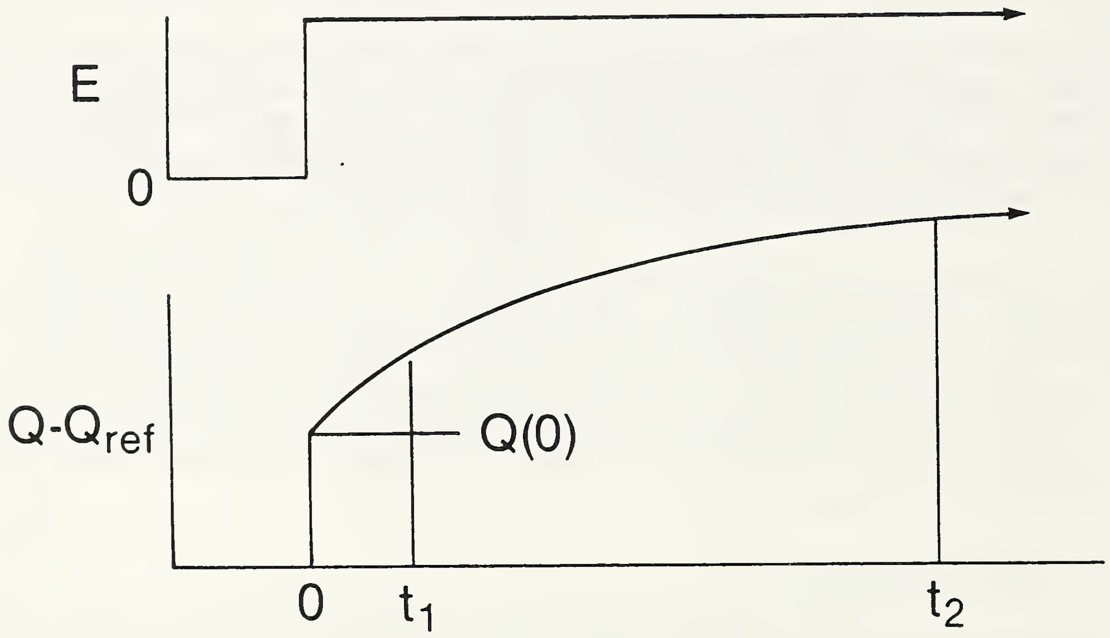

Fig. 1. Applied field (top curve) and measured charge response for the time-domain measurement. Q(0) is the measured charge immediately after the application of the field. The times t_1 and t_2 are the shortest and longest times of measurement and also define the frequency range of the measurement.

$$C'(\omega) = \int_0^\infty \frac{dC}{dt} \cos \omega t \, dt ,$$

$$C''(\omega) = \int_0^\infty \frac{dC}{dt} \sin \omega t \, dt . \tag{9}$$

The term C″ can be defined as $C'' = G(\omega)/\omega$ so that

$$\epsilon^* = C'/C_g - iC''/C_g = C(\omega)/C_g - iG(\omega)/C_g . \tag{10}$$

The equations clearly apply to the entire frequency range, so that a single step response measurement is equivalent to measuring the entire, continuous frequency range. The measurement advantage is extremely large since one can only take a constant frequency measurement one frequency at a time with the excitation applied for at least several cycles.

The equations, to cover the entire range, assume an equivalent coverage in time with short time corresponding to high frequencies and long times to low frequencies. In practice, the finite response times of the circuitry will set a limit, t_1, on the shortest time observable while either instrumental limitations or even a desire just to end the measurement will set another limit, t_2, as the longest time. These limits act to set a practical limit to the frequency range that can be covered. For t_1 equal to 10 μs, as in the Spectrometer, a maximum frequency of 10 kHz is obtained. Even more significant is the result that t_2 corresponds to 0.3 times the period of the lowest observable frequency. Thus a measurement of 300 s results in a frequency coverage of 0.001 Hz to 10 kHz, or seven decades. Furthermore, since a discrete FFT is not used[15], any frequency in the covered range is obtainable.

Just as important as the frequency data are the time data. The value of dC(t)/dt for t = 1 min is the proper definition of the ASTM conductivity. Therefore comparisons can be made not only with other loss measurements, but also any DC conductivity measurements. It should be noted from this discussion, that the an apparent determination of DC conductivity can be very misleading if dielectric loss is present, as is usually the case for polymeric materials. For typical reactor cable insulation, a measurable change in value for DC conductivity may or may not correspond to a change in ionic conductivity upon irradiation[16]. As a result, the interpretation of the data as only due to induced conductivity can be erroneous. This report will demonstrate this point clearly.

The equipment needed to carry out this measurement is outlined in Figure 2. Two matched, high speed precision step generators, with a rise time of less than 3 μs and with a settling time of ≈10 μs, apply a 100 V step across the sample and a lossless reference. The difference signal is followed with a charge amplifier with a response time of 1μs and a bias current of <30 fA. The signal is then digitized and clocked into a computer using a clock with programmable rates from 5 μs to 10 s. The data are then transformed and displayed. The shortest time corresponding to the 10 μs settling time corresponds to a frequency of 10 kHz while terminating the measurement at 300 s permits extraction of frequency data to a frequency as low as 10^{-3} Hz.

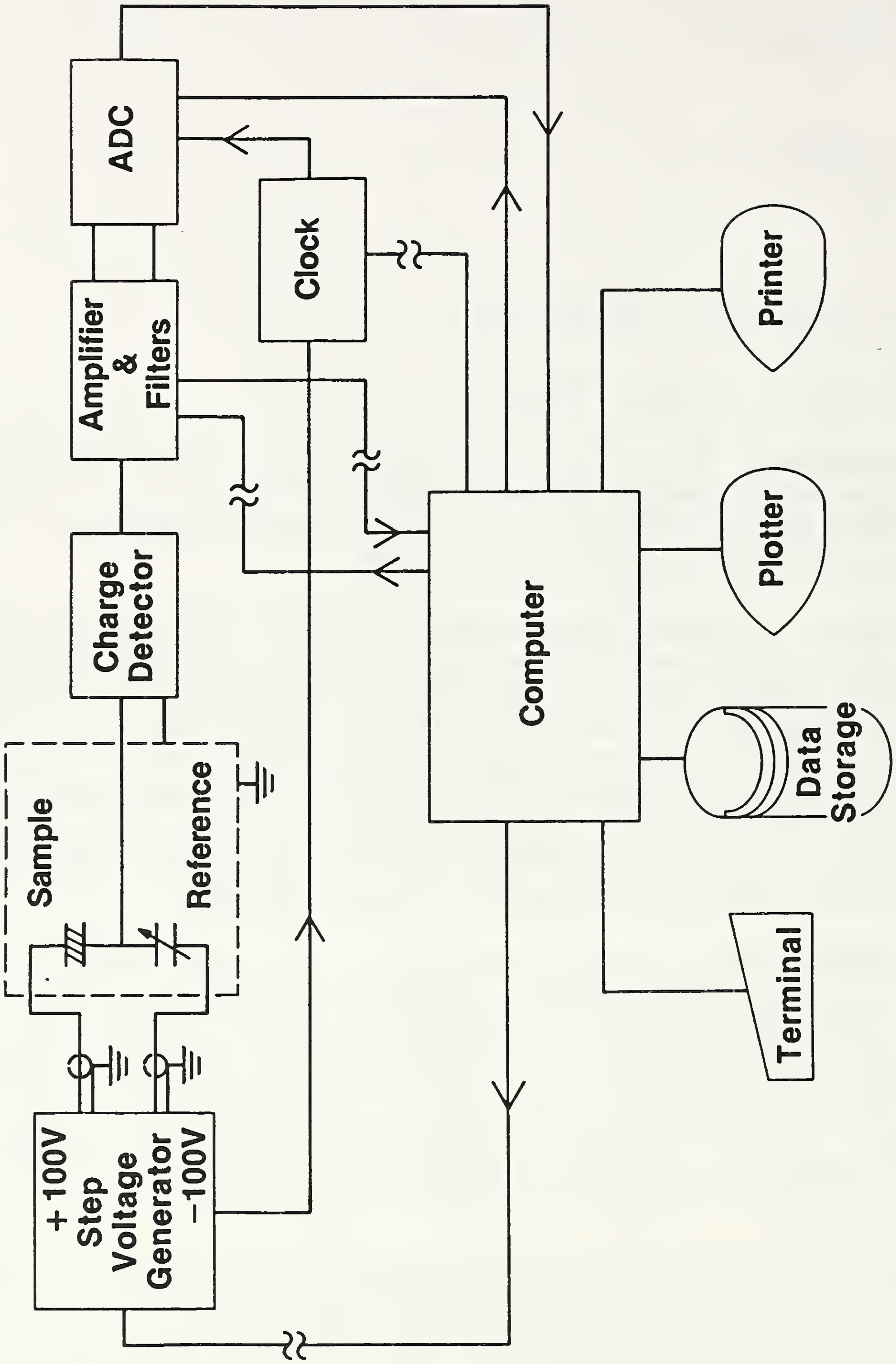

Fig. 2. Block diagram of the Dielectric Spectrometer. The breaks in the data lines, the directions shown by arrows, represent optical isolators.

Sample Requirements

Since the emphasis in this report is on cables, the sample geometry for this study was chosen so that actual cable samples could be used. For a coaxial cable, the insulation between the shield and the center conductor is clearly the most important insulation in the system, as the jacket is for protection and separation of ground planes. This allowed the use of the measurement configuration shown in Figure 3. The center conductor is connected to instrumental low using a shielded, coaxial lead. The cable shield is connected to instrumental high using an ordinary clip lead. The entire cable was enclosed in a shielded box such that the box was connected to the detector ground while the shielded lead from the generator has its ground ended at the box without contact. This arrangement ensured that no ground loop currents were included in the measurement.

Only the impedance between high and low is measured and not any impedances to ground. Any stray coupling, including that involving the jacket, was not measured. Also, if it proved necessary, guard rings could be applied at the ends of the cable by stripping sufficient braid away from the inner insulation. These rings can intercept any surface conductivity in the cable. For the results reported here, no effects of surface conductivity were ever found and most samples were used unguarded.

For those cables that did not have an outer braid and jacket, conductive paint was applied to the exterior so that a center electrode and two guard rings were formed. This then allowed the cable to be measured exactly in the same way as the coaxial cable samples. In order to avoid shorting the outer electrode, a sheet of fluorocarbon polymer was placed under the sample. The lead shielding insured that this insulating sheet was not part of the measuring path.

The length of the sample was such that the sample capacitance was always greater than 20 pF. This value of capacitance was large enough so that, except for the very lowest loss, unaged samples, instrumental noise and resolution were not limiting factors. For the cable samples used in this study, the longest lengths were about 35 cm as measured by the length of the outer electrode, which controlled the effective electrical length.

The geometric capacitance in picofarads was computed from the sample length and the diameters of the inner and outer electrodes, assuming coaxial cylinder geometry with the formula

$$C_g = (0.5563\ l)/\mathrm{Ln}(d_1/d_2)\ , \tag{11}$$

where l is the electrode length in cm, d_1 is the diameter of the outer electrode and d_2 is that of the inner. For the samples used in this study the major uncertainty in C_g was in the determination of d_2, especially for those cables that had small diameter braided center conductors. For the purposes of this report, the values of C_g are probably accurate to 5% and should not affect any conclusions made.

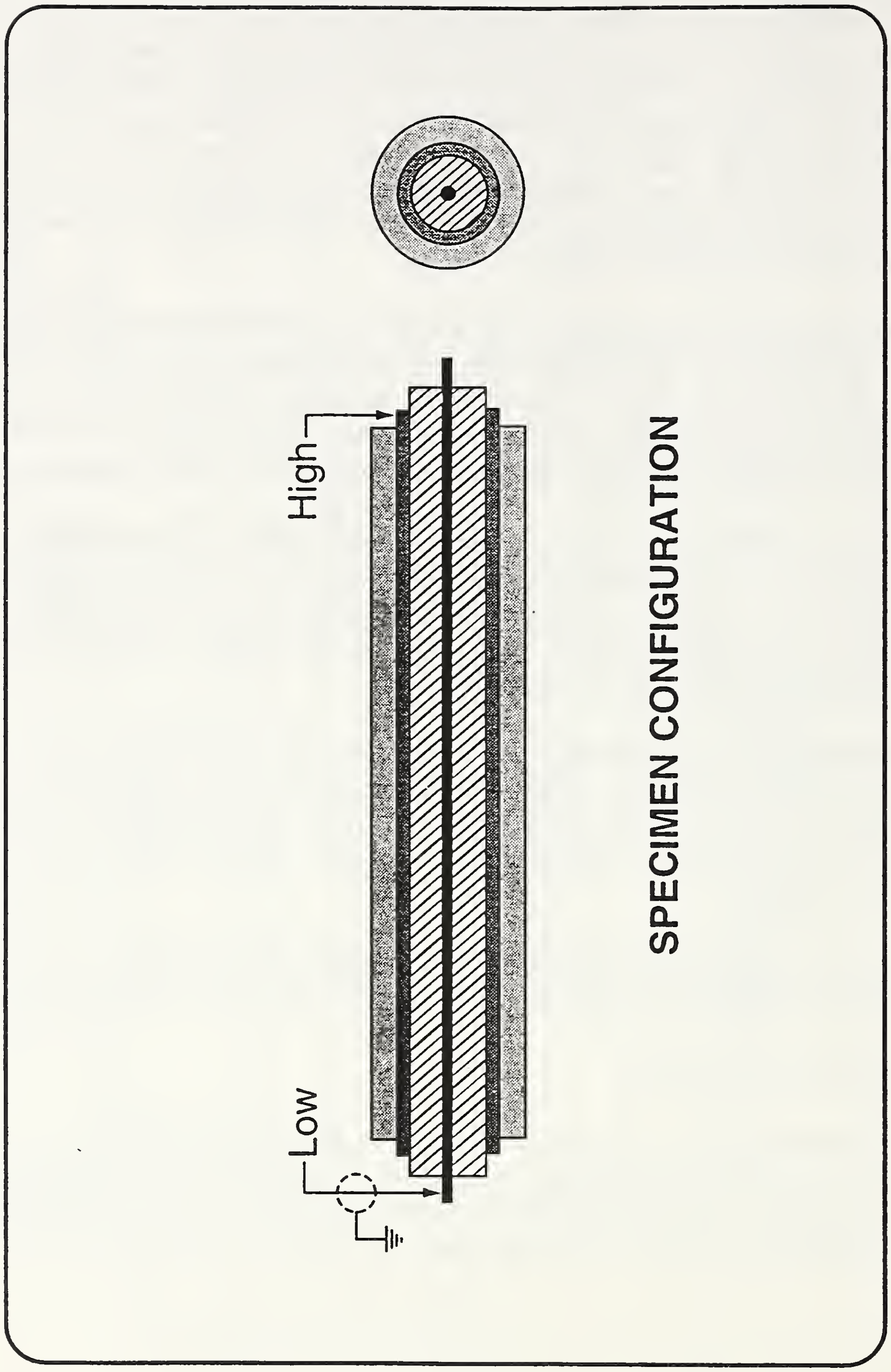

Fig. 3. Typical cable sample configuration used in this report. The braid and center conductors were used to define a coaxial sample geometry. The guard rings shown outside the outer braid and connected to ground were optional.

SAMPLES

The greatest difficulty encountered in this study was in obtaining a systematically aged set of samples so that a progressive study could be made. While this difficulty has set a definite limit as to the conclusions of this report and leaves questions open, it will be seen that a sufficient range of samples were obtained to state that dielectric data can clearly be used to follow the aging of reactor cable insulation.

All the cable samples are of the type that have been used in reactors or are prototypes of those that could. Except for the cable samples recovered from the Shippingport reactor, the samples examined in this study were all subjected to accelerated aging protocols. Only one of them, the Sandia coaxial cable, was subjected to a simulated LOCA and it did not fail.

All the measurements shown in this report were carried out at 50°C. For some of the samples, measurements at other temperatures were carried out, only to confirm the results presented here. The chosen temperature optimized total measurement time and allowed a firm comparison to be made between different sets of measurements. Any important sample considerations will be presented with the discussion for that set of samples. Table 1 presents a summary of all the samples with the values of ϵ measured at 10 kHz.

Shippingport

Two samples were obtained from the decommissioned Shippingport reactor. Both of them were from the same lot of RG149/U cable manufactured by the Times Wire and Cable Company. This form of cable is insulated with polyethylene, has a vinyl jacket, and is no longer certified for use in reactors. At Shippingport, these cables were acknowledged to have a limited lifetime and were replaced on a regular basis every three years.

One of the samples was recovered from the control room which was not exposed to the reactor environment. It served as the baseline control. The other piece was recovered from inside the reactor confinement. It was clearly exposed to radiation but, as the records were not available, it is not known as to how long the cable was in service. The vinyl outer jacket showed a tendency to crack and the polyethylene insulation had a light yellow color.

Pieces of both cables were measured at 50°C over the frequency range of 10^{-2} to 10^{4} Hz. The results of the dielectric loss measurements, expressed as $\tan \delta = \epsilon''/\epsilon'$ are shown in Figure 4. The measured values of $\tan \delta$ for the unirradiated cable are quite comparable with those observed in any fresh sample of coaxial cable using polyethylene as a dielectric. The irradiated cable, on the other hand, shows enhanced loss at both the high and low ends of the frequency spectrum. The dramatic increase in $\tan \delta$ below 1 Hz is consistent with a conduction mechanism since the increase is proportional to $1/\omega$. The much smaller rise above 1 kHz will be seen upon comparison with other data to be the first appearance of a loss process that occurs upon irradiation of aliphatic hydrocarbon polymers.

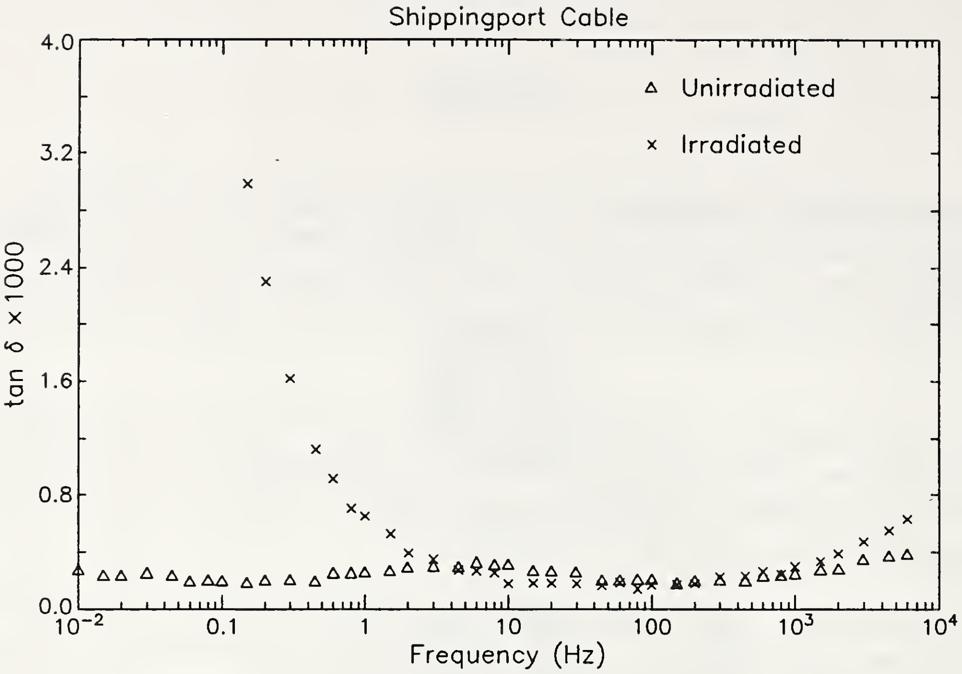

Fig. 4. The dielectric loss, tan δ, for two samples of coaxial cable recovered from the Shippingport reactor. The unirradiated sample was from the control room while the other was from inside the containment.

Sandia

XLPO

Several different samples were donated to us by Mark Jacobus of Sandia National Laboratory. There were pieces of single wire insulation labelled as being made of XLPO (cross-linked polyolefin), both unaged as well as exposed to 25 Mrad. These samples were colored and pieces of black, red and white were supplied. There were also samples of a coaxial cable of XLPE (cross-linked polyethylene) in the unaged state as well as aged at 100°C with doses of 36 and 65 Mrad. The data for these samples are displayed in Figures 5-12.

The XLPO wire insulation was supplied as only the insulation with no center conductor. The mechanical properties were measured for this set of samples[17] and they showed little change for this particular dose. There was no apparent change in appearance for these samples. For these samples, electrical measurements were carried out by inserting 14 Ga. copper wire in the samples and electrodes painted on the outside. The dielectric constant of 2.6 at 50°C and 10 kHz for all samples is clearly too high for an aliphatic hydrocarbon polymer and suggests the presence of a mineral filler.

The loss data for all three colors of polyolefin, both unaged and aged are shown in Figures 5, 7, and 9 as a function of frequency. In order to interpret the data more easily, the same data are shown replotted in Figures 6, 8, and 10 with frequency being replaced by the real part of the complex dielectric constant, ϵ', as the independent variable. This complex plane plot, the Cole-Cole plot, often separates different dispersion regions of the loss data more clearly without distorting the accuracy of the data. Also, since ϵ' decreases with increasing frequency monotonically, frequency becomes a running parameter along a displayed curve with frequency decreasing left to right. In all the data displayed in this report, the frequency decades, starting at 10 kHz on the left, are marked on the plots so that the data can be more easily compared. The curves shown are not interpolations but are the actual data.

An important consideration for the discussion in this report is that in the Cole-Cole plot a conductance is represented as a vertical line since there is no change in ϵ'. A dipolar mechanism, on the other hand creates a closed, smooth curve intersecting the real ϵ' axis at both the limiting values for high and low frequency. In practice, as relaxations are rarely completely separated, the closed curves are not completed before another relaxation dominates, but inflection points usually clearly reveal the relaxations. In all the plots shown in this report, when multiple curves are displayed in the complex plane, they are arbitrarily displaced along the horizontal axis for clarity. The starting, 10 kHz value for ϵ' for each curve is given in Table 1.

For the XLPO, insulation of all three colors behaved similarly with an insufficient number of samples available to indicate as to whether the small differences between the colors are significant. The unaged samples all showed a large increase in loss with decreasing frequency. The Cole-Cole plots show that this loss can not be represented by a simple conductivity since the curves are distinctly not vertical but have a finite slope as if the loss were the beginning of a major, very low frequency dielectric loss region. However, the apparent magnitude of the dispersion is indicative of an interfacial polarization, probably induced by the filler.

Upon irradiation with 25 Mrad, all three colors of XLPO showed a major change in behavior. The apparent low frequency loss decreased, in strong contrast to the expectation that it might increase from radiation induced conductivity. The data obtained on these samples are insufficient to state this as

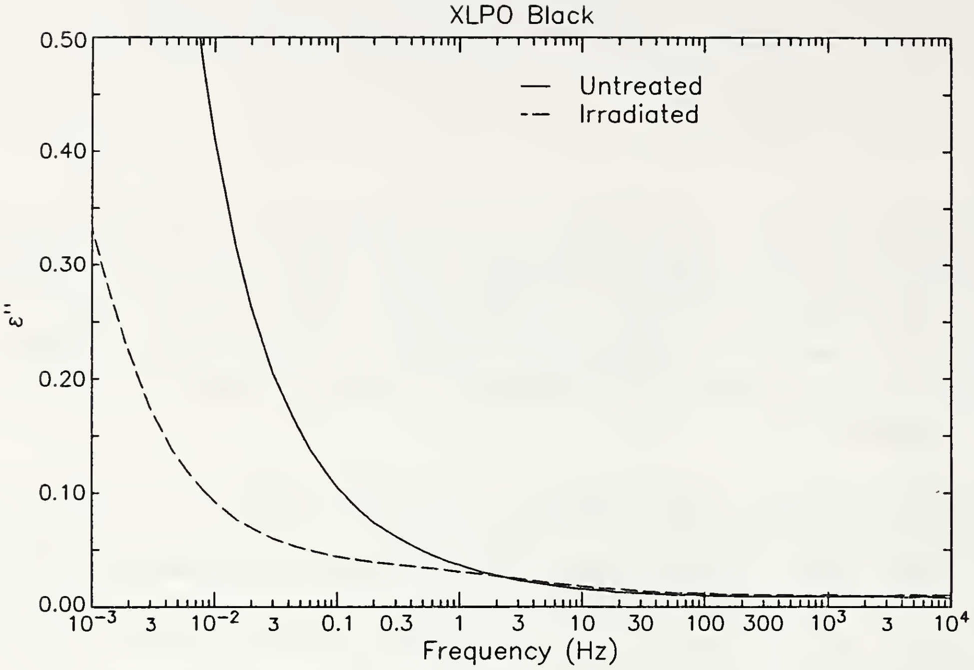

Fig. 5. The loss index, ϵ" for black XLPO wire insulation. The irradiated material was exposed to 25 Mrad. Note the decrease in loss upon irradiation at the lowest frequencies, but the crossover above 1 Hz.

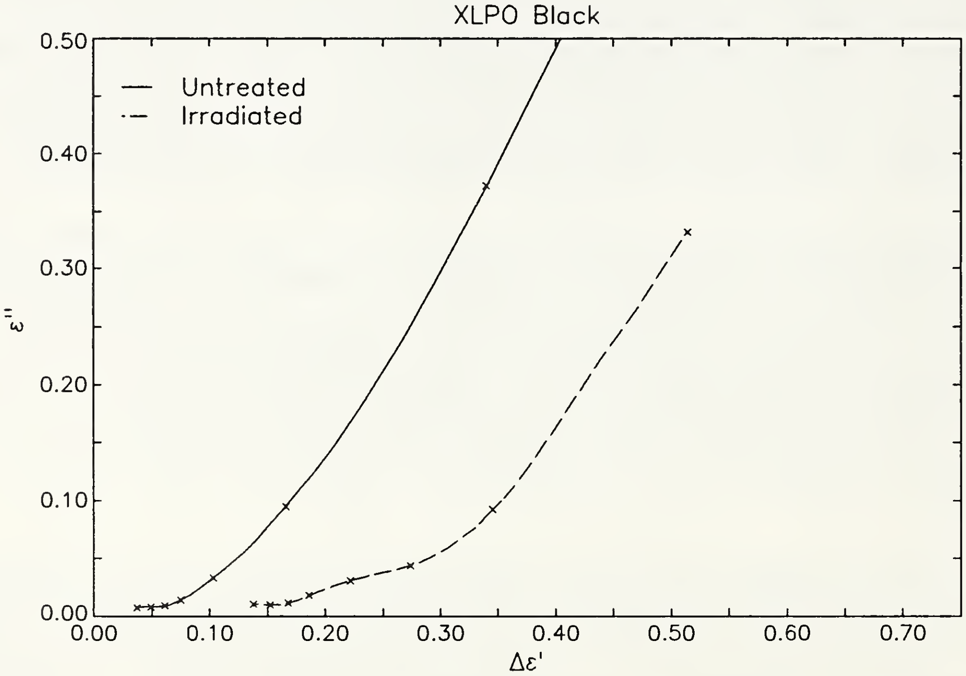

Fig. 6. The data for black XLPO replotted in the complex plane vs ϵ'. The crosses mark the frequency decades. The data are arbitrarily displaced along the real axis. The shoulder at Δϵ' = 0.25 is dipolar loss.

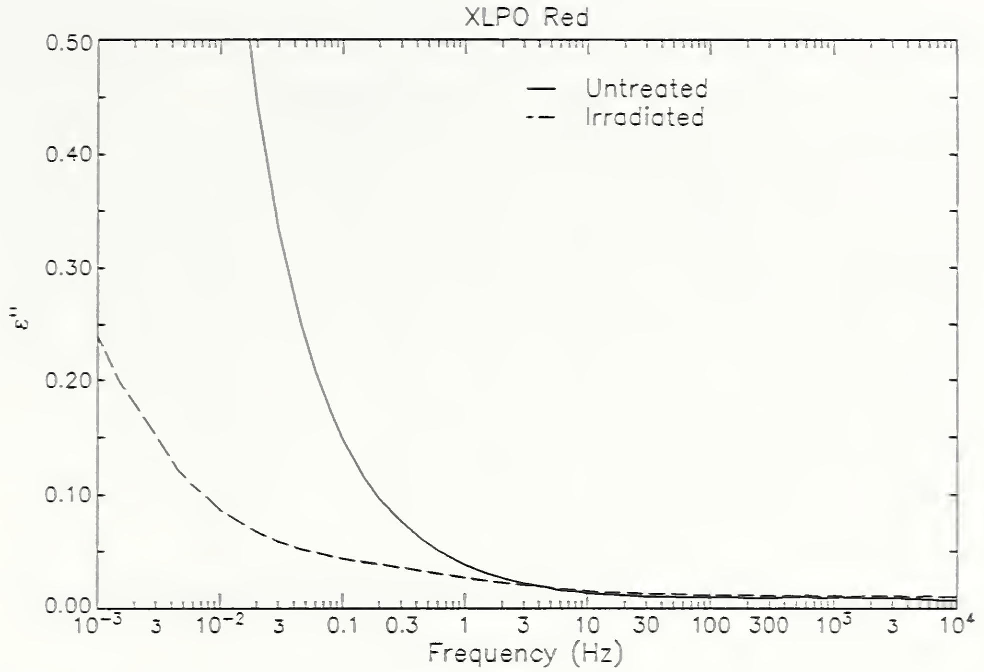

Fig. 7. The loss index, ε" for red XLPO wire insulation. The irradiated material was exposed to 25 Mrad. Note the decrease in loss upon irradiation at the lowest frequencies, but the crossover above 1 Hz.

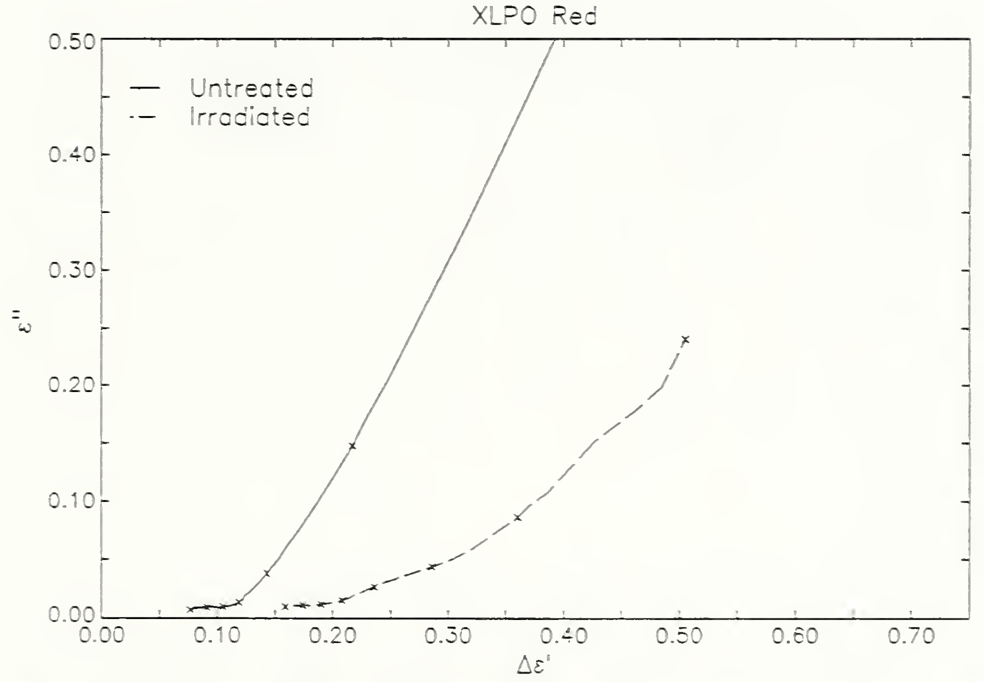

Fig. 8. The loss index, ε" for red XLPO replotted in the complex plane vs ε'. The irradiated material was exposed to 25 Mrad. The incease in loss above 1 Hz appears as a shoulder at Δε' = 0.25.

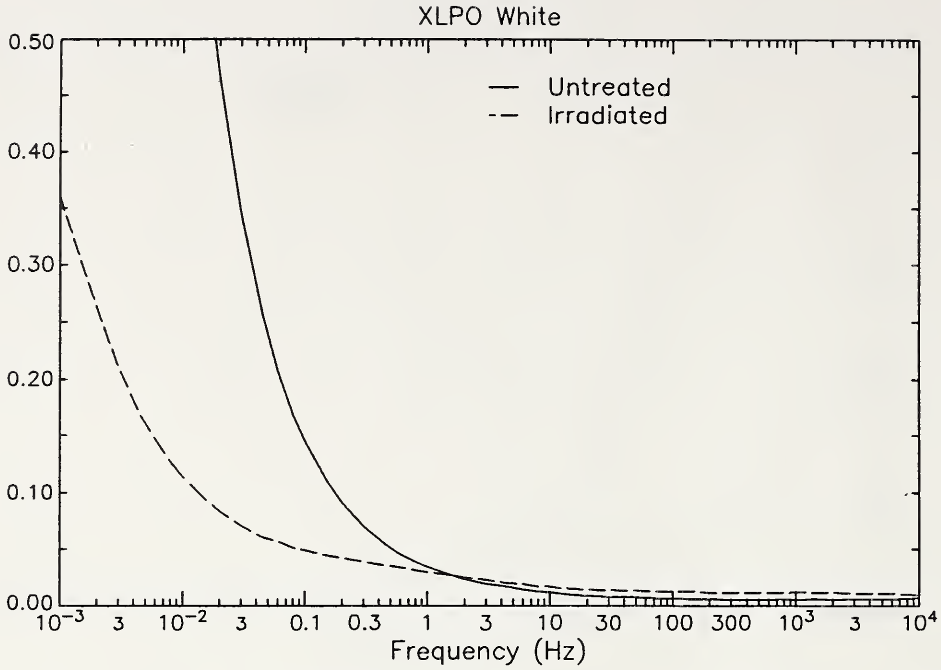

Fig. 9. The loss index, ϵ'' for white XLPO wire insulation. The irradiated material was exposed to 25 Mrad. Note the decrease in loss upon irradiation at the lowest frequencies, but the crossover above 1 Hz.

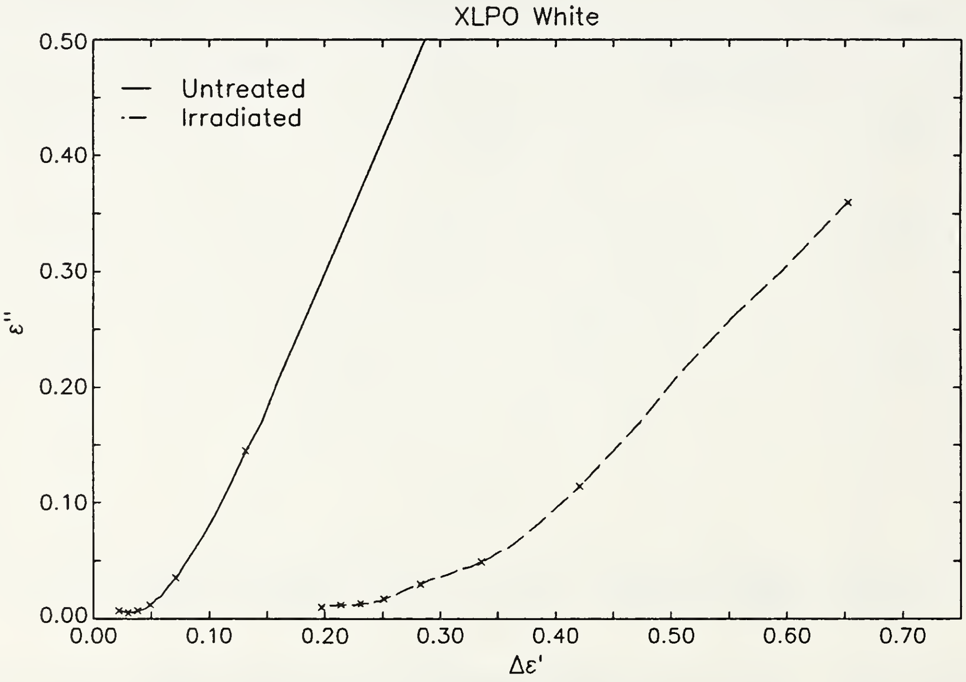

Fig. 10. The loss index, ϵ'' for white XLPO replotted in the complex plane vs ϵ'. The irradiated material was exposed to 25 Mrad. The incease in loss above 1 Hz appears as a shoulder at $\Delta\epsilon' = 0.25$.

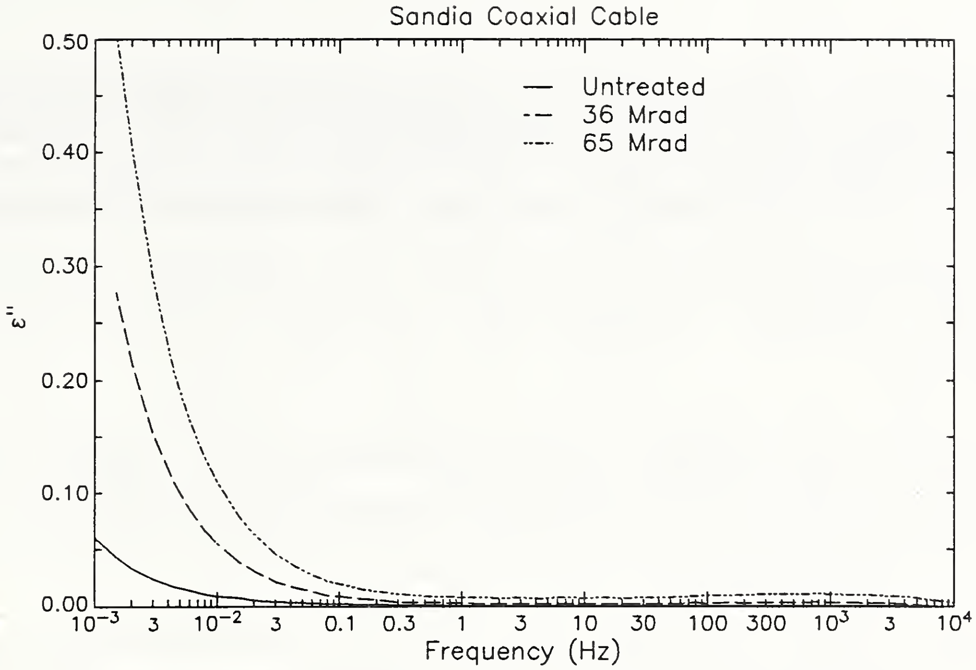

Fig. 11. The loss, ϵ'', for the Sandia coaxial cable as a function of dose.

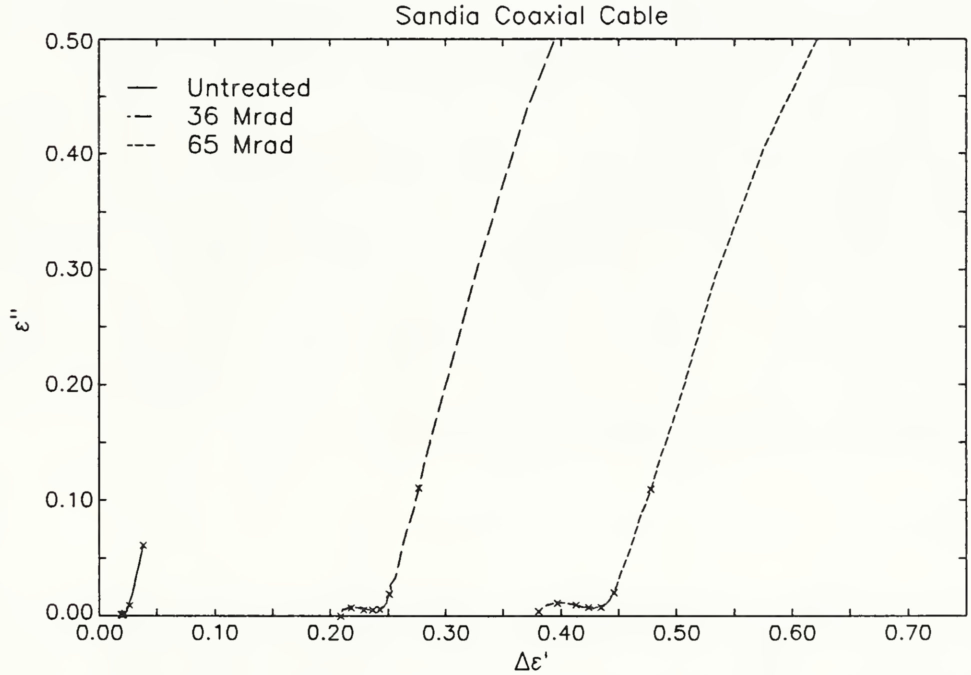

Fig. 12. The loss data for the Sandia coaxial cable replotted in the complex plane. Note the increasing dipolar loss region with dose that appears as a toe to the loss curves.

a fact as it was not pursued and there could be, instead, a shift to lower frequencies. More significantly, careful examination of Figures 5, 7, and 9 shows an increase in loss around 30 Hz. This loss in Figures 6, 8, and 10 is revealed as an easily observable shoulder indicative of a new loss process appearing upon irradiation. This loss will be seen to be a characteristic of radiation aging in all the polymers examined in this report.

Coaxial Cable

The other cable samples supplied by Sandia were pieces of a coaxial cable certified for use inside reactor confinement. It was the first set of samples studied with more than one dosage. The available samples were unaged. exposed to 36 Mrad at 100°C and exposed to 65 Mrad at 100°C. The data for these samples, measured at 50°C as for all other data, are shown in Figure 11 as a function of frequency and Figure 12 in the complex plane.

The cable showed little visual signs of aging except for a reddish color in the translucent central insulation. From the lack of opacity and value of the dielectric constant, there was no evidence of a mineral filler as in the previous samples. The loss values for the unaged sample are low but are somewhat higher than those found in a good low loss cable.

Upon radiation aging, there is a clear increase in loss at frequencies below 0.1 Hz. While there is clear curvature in the complex plane plot, the data are not complete enough to distinguish between a dipolar loss process and possible interfacial effects involving the cable conductors.

More revealing is the increased loss at frequencies above 0.1 Hz that is clearly a dipolar loss mechanism and that grows with increasing dose. While this increased loss is hard to see in Figure 11, it is clearly visible in Figure 12 where the relaxation process is seen on the left side of the curves for the aged samples. In this Cole-Cole plot, the amplitude of the relaxation is given by the difference between the high and low values for the extrapolated intersections with the real axis. For these sets of data, this extrapolation is quite clear with an estimated value for the amplitude for the 36 Mrad sample of 0.03 and for the 65 Mrad sample of 0.05. It should be noticed that this relaxation takes place at nearly the same range of frequencies as that found in the XLPO samples. Also, the unaged sample does not have this loss process, again similar to the case found in the XLPO samples.

EDF Cables - First Series

The first cables obtained from Electricitè de France (EDF) was a set of five test cables that had previously been studied for aging. They were available in the original unaged state, exposed to 85°C thermal aging, exposed to 25 Mrad, and exposed to both 25 Mrad and 85°C. The construction of each cable type was different, having different shielding configurations. Several had solid metal foil wraps and several had a semiconductive layer applied. They also varied as to their characteristic impedance. None of these differences is believed to be significant and no obvious correlation was noted.

The only important factor was the nature of the insulation. Cables 1-3 were cross-linked polyethylene while Cables 4 and 5 were ordinary polyethylene. Cables 1 and 2 had fire retardants added. These differences had strong consequences for the initial properties as well as the response to aging. The data obtained on these samples are displayed in Figures 13-22.

Cable 1 and Cable 2, Figures 13-16, both had flame retardant added to a cross-linked polyethylene insulation. The retardant was different in the two cases but unspecified. Both cables had excess loss in the unaged state, especially Cable 2, compared to unaltered polyethylene. Cable 2 did show evidence of additive migration in the unaged samples. There was no visual evidence of radiation aging in either cable. The presence of the retardants in both cables seems to be an inhibitor of radiation damage at the dose of 25 Mrad that was used. The decrease in loss upon thermal treatment found in both cables could be the result of the removal of a volatile ingredient. Especially in Cable 2, where the radiation induced conductance is quite low, there is evidence for a new loss peak appearing upon irradiation.

Cable 3, Figures 17, 18, a cross-linked polyethylene cable, showed a clearer progression with aging. This cable did show some enhanced loss upon exposure to thermal aging. The loss region that has been found to appear upon irradiation is seen most clearly as a shoulder in the data for the heated and irradiated sample in Figure 18 and the enhanced loss at 1 Hz for the same sample in Figure 17.

Cables 4 and 5 were both polyethylene cables that were not cross-linked. The data are shown in Figures 19-22. The original cables, even upon heat treatment, were extremely low loss and data for these samples are not readily visible in the plots shown. The single points shown in Figures 20 and 22 represent the entire frequency range for the samples not irradiated. However, they both showed the most extensive changes upon irradiation. The rapid rise in loss in Figures 19 and 21 correspond to the steep, nearly vertical traces in Figures 20 and 22. This excess loss is clearly the result of radiation induced conductivity from degradation. The presence of low molecular weight species resulting from irradiation was noted by the presence of a waxy odor upon heating to 50°C. The loss region found earlier is just observable in the frequency plots as a region just above the frequency axis and as a small toe in the complex plane plots.

EDF Cable - Prototype series

Dielectric Data

The only set of samples that had a relatively wide range of conditions applied to them was for a prototype cable supplied by EDF in France. There were 19 samples furnished with varying thermal treatments and with radiation doses of not only 25 Mrad but also 85 Mrad. The range of samples is given in Table 2. The cable was a high impedance (115 Ohm) cable with an insulation of EPDM (ethylene propylene diene modified) rubber that had an applied conductive polymer layer underneath the braids. The cables exposed to the highest temperatures displayed clear evidence of thermal degradation. The outer jackets of a chlorinated polymer were hard, brittle and darkened while the EPDM insulation had a green coloration typical of copper compounds. The tinned surfaces of the braid and the inner conductor were clearly corroded and in several of the cables, the inner conductor was found to be broken. These observations are noted in Table 2.

For those samples that had a broken inner conductor, capacitance measurements indicated that the breaks were approximately in the center of the sample. For these, both ends of the inner conductor were exposed and connected by a shielded cable whose braid was grounded. This configuration prevented any contribution from the joining cable entering into the measurement, yet allowed full use of the cable

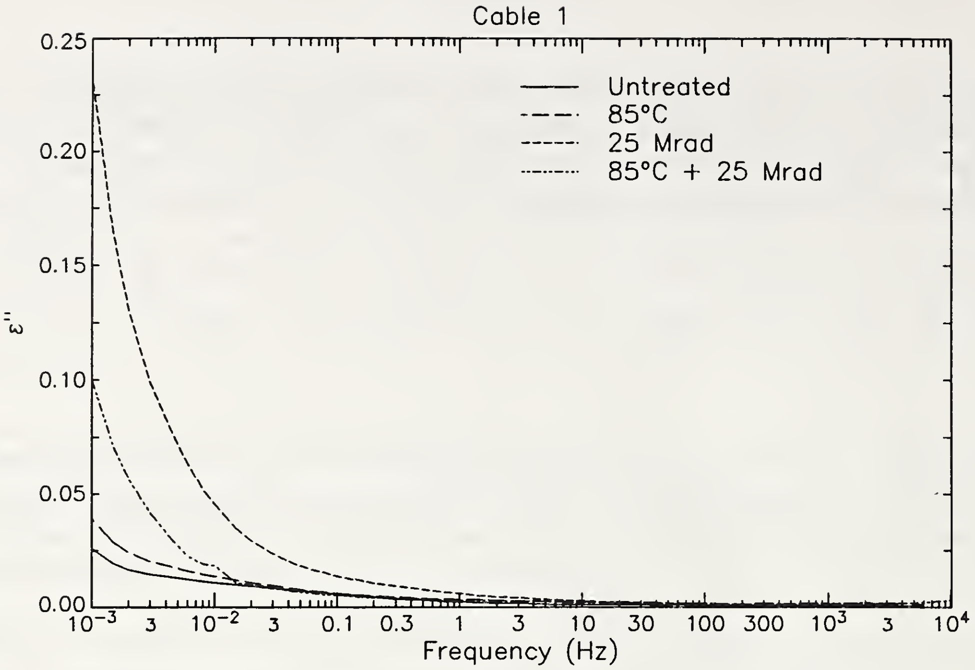

Fig. 13. The loss data for EDF Cable 1 as a function of treatment.

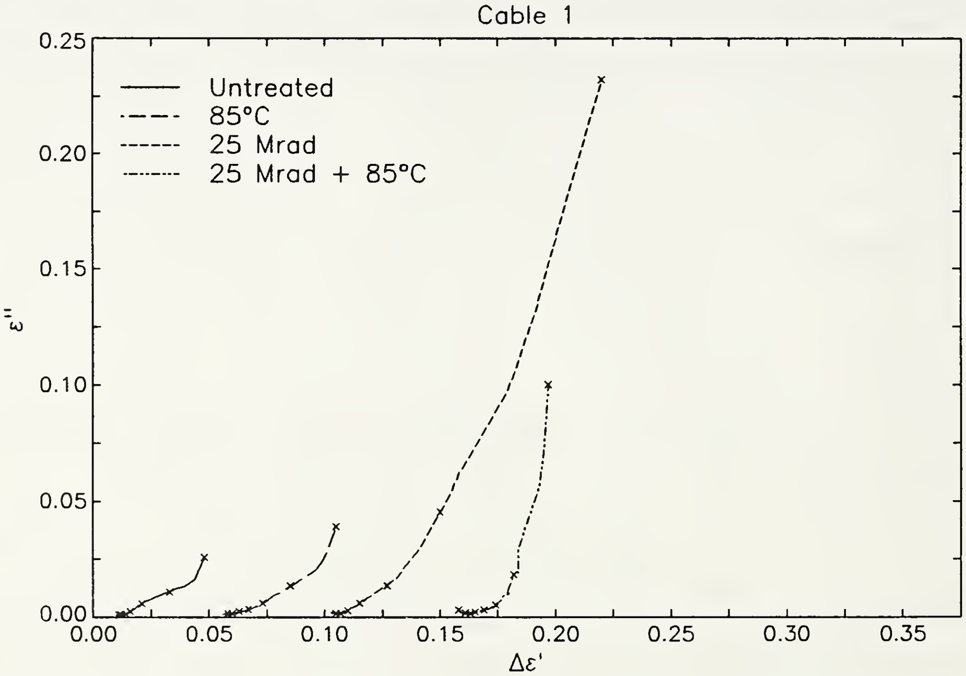

Fig. 14. The loss data for EDF Cable 1 replotted in the complex plane. The break in the curve for the 25 Mrad + 85°C sample is not real.

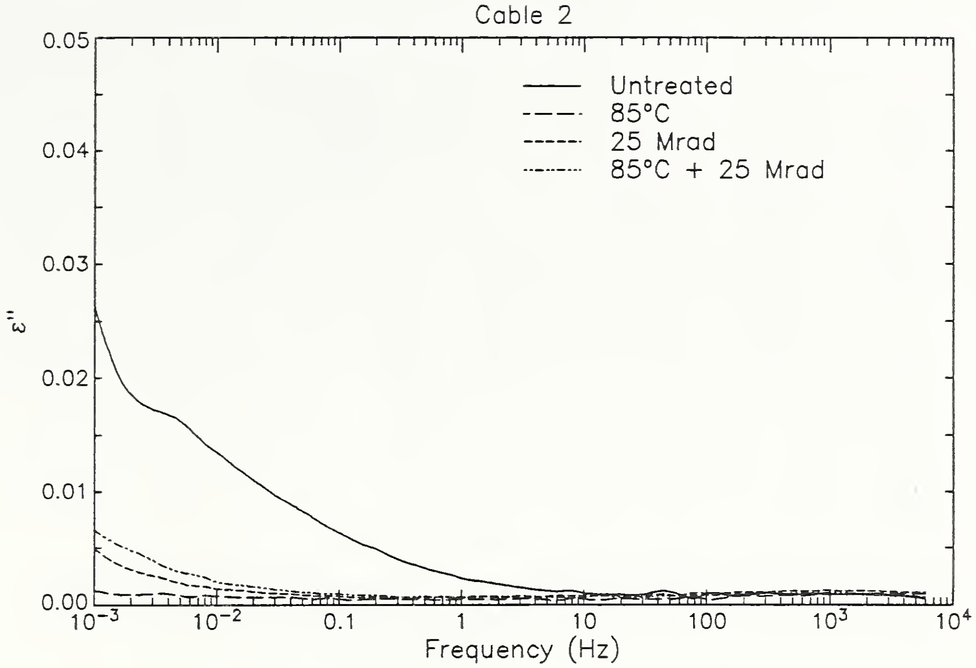

Fig. 15. The loss data for EDF Cable 2 as a function of treatment. The shoulder at 10^{-2} Hz is due to the flame retardant.

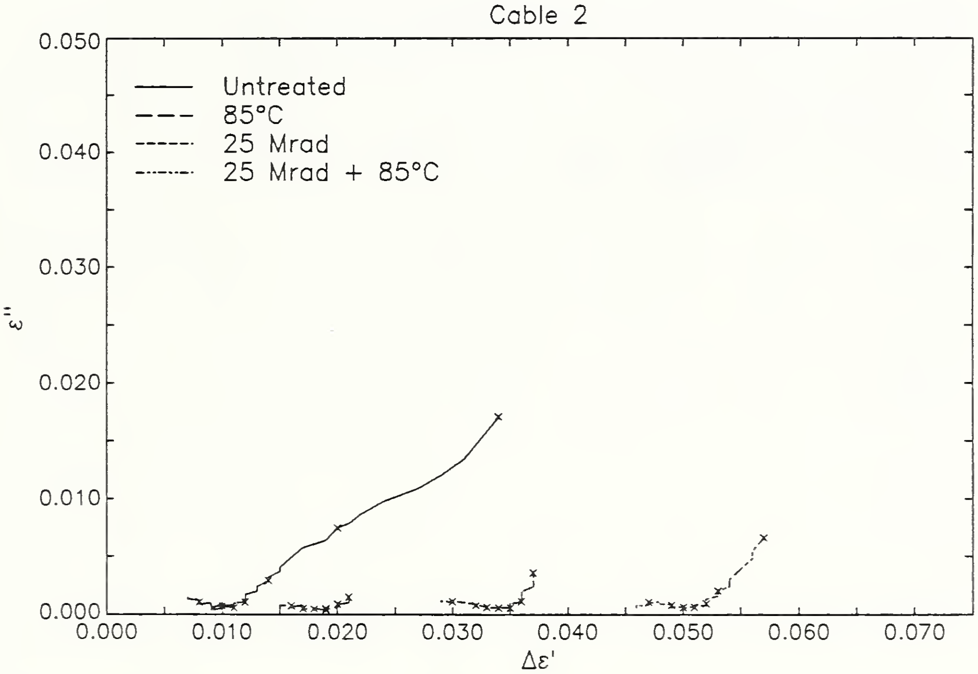

Fig. 16. The loss data for EDF Cable 2 replotted in the complex plane. While the loss decreases upon treatment, there is a new loss region upon irradiation seen at $\Delta\epsilon'$ of 0.030 and 0.050.

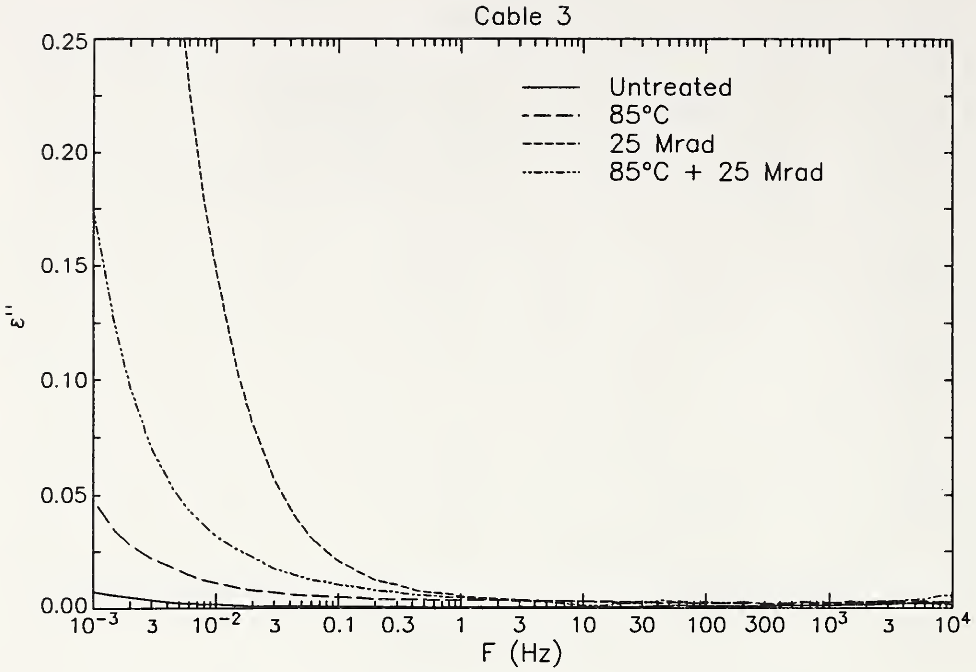

Fig. 17. The loss data for EDF Cable 3 as a function of treatment. There is an excess of loss above 1 Hz upon irradiation.

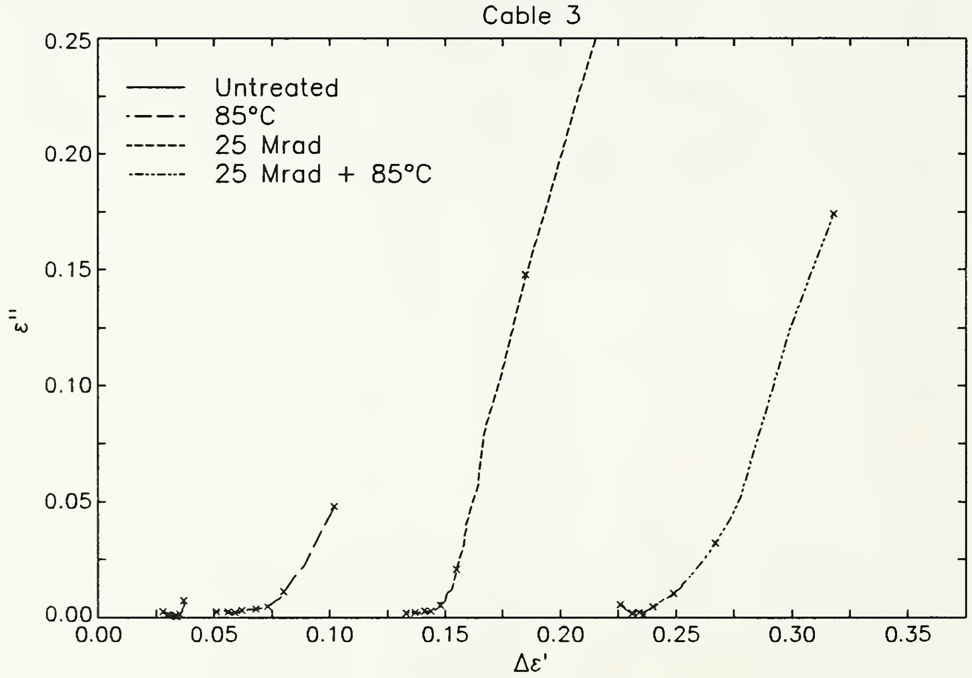

Fig. 18. The loss data for EDF Cable 3 replotted in the complex plane.

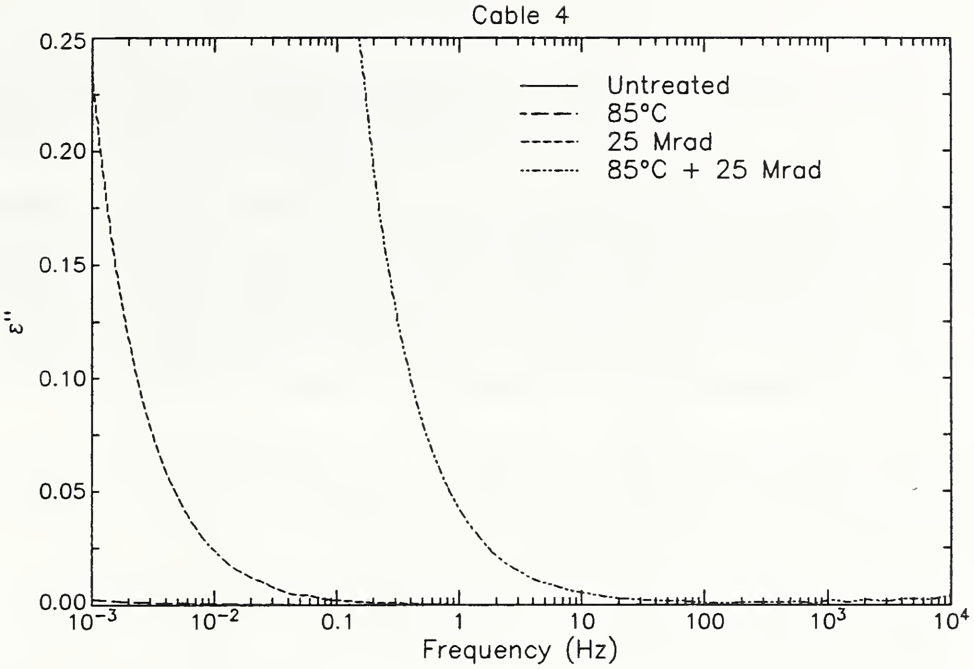

Fig. 19. The loss data for EDF Cable 4 as a function of treatment. The dat for the untreated and heated samples are along the frequency axis.

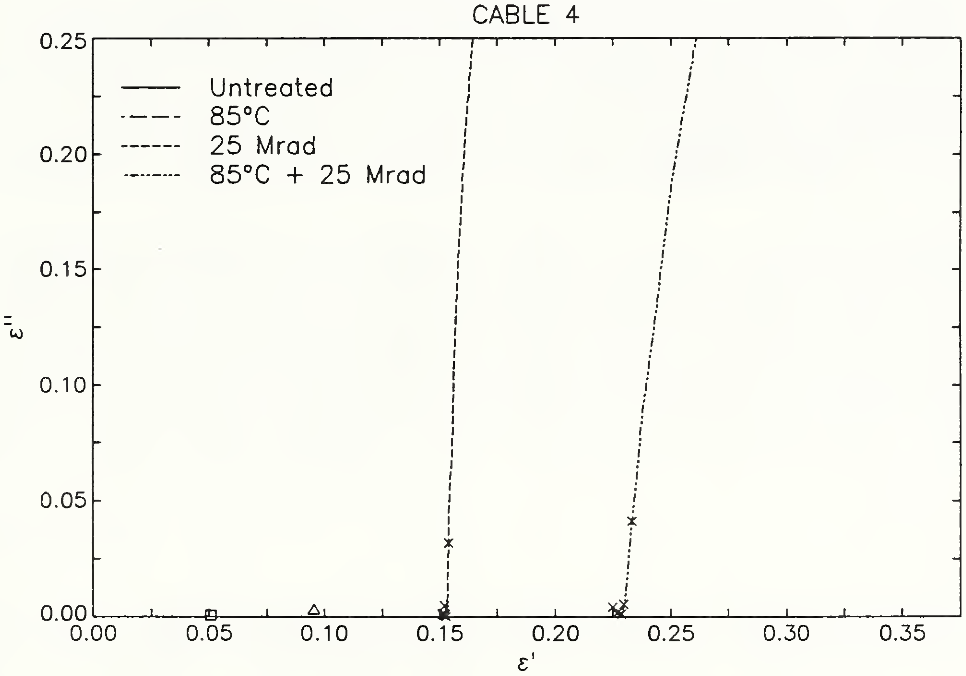

Fig. 20. The loss data for EDfF Cable 4 replotted in the complex plane. The unirradiated and heated samples are under the single plotted points. The near vertical lines for the irradiated samples are due to conduction.

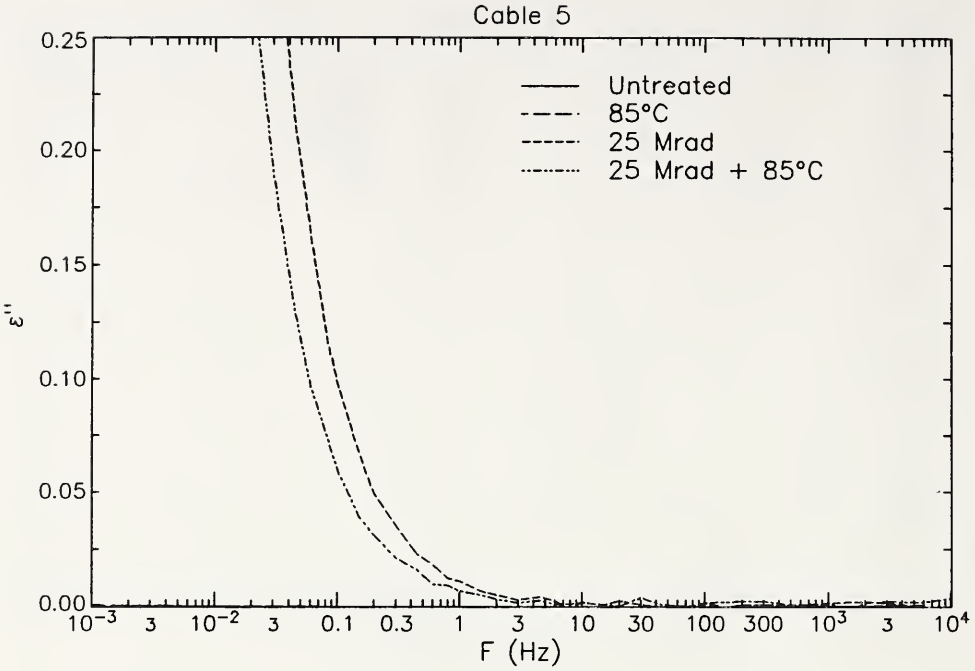

Fig. 21 The loss data for EDF Cable 5 as a function of treatment. The untreated sample is along the frequency axis.

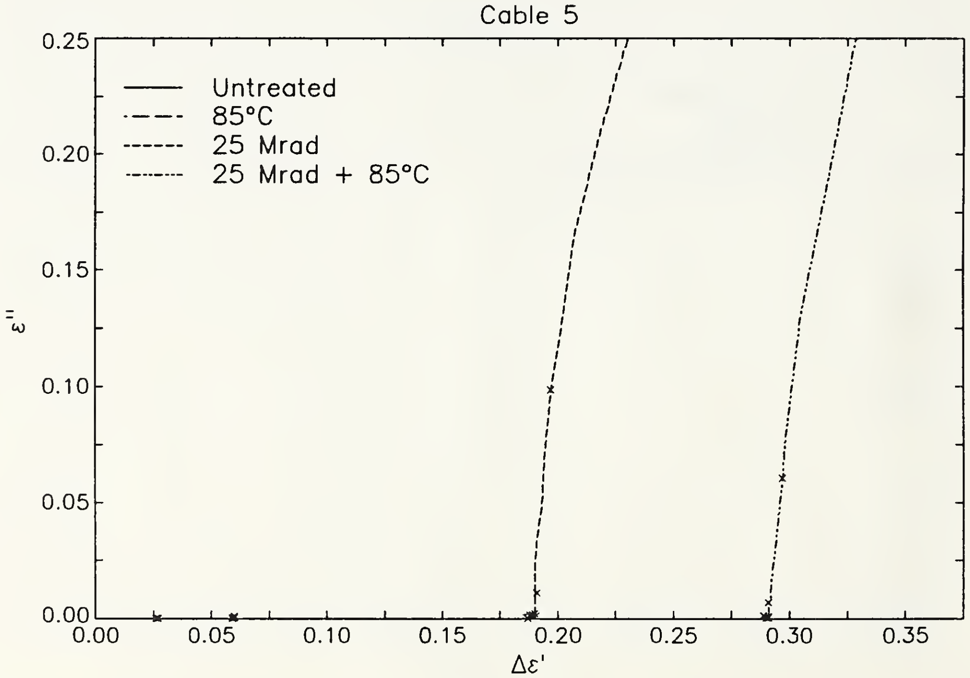

Fig. 22. The loss data for EDF Cable 5 replotted in the complex plane. The unirradiated sample data are under the single plotted point.

insulation. Also, since there was no free surface of the insulation present, the full sample could be measured without concern for superfluous surface conductivity.

The data for the results obtained on the 100°C aged samples are shown in Figures 23-28. None of these samples showed much in the way of visual degradation except for the high dose, long thermal exposure samples which had the originally white outer jacket show a slight yellowing. The dielectric data for all these samples are quite similar and serve to illustrate the reproducibility of the results. For reference, the untreated sample is plotted in Figures 23 and 24 but, on the scale needed to represent the other data, only the single cross in Figure 24, representing the entire spectrum, near the origin is visible.

For all the 100°C aged samples, thermal aging did induce a small amount of loss that changed slightly with increased exposure. No major relaxations are resolved in the frequency range of 10^{-3} to 10^{4} Hz. For the irradiated samples, on the other hand, a progressive increase in loss is clearly evident on progressing from 25 Mrad to 85 Mrad. Irradiation clearly induces a dipolar loss process, that in these samples is visible as shoulders even in the frequency plots below 1 Hz. At 85 Mrad the dipolar loss has increased in magnitude and shifted somewhat to higher frequencies. This can be noted in the complex plane plots by the size of the shoulder.

The samples exposed for different periods of time at 135°C clearly were subject to thermal degradation. The data for these samples are shown in Figures 29-34. These samples were clearly heated above the degradation temperature for the outer jacket and the subsequent liberation of HCl. The deepening color of the outer jacket, even upon just thermal aging, the increased corrosion apparent on the conductors and the increasing likelihood of conductor breaks demonstrates this point. For these samples, thermal aging alone was sufficient to induce substantial dielectric loss. The steep slopes in the complex plane plots, Figures 30, 32, and 34 for the unirradiated samples is consistent with an induced ionic conductivity. These samples seem to indicate a small decrease in loss with time, perhaps from a slowing of the degradation reaction and a volatilization of the reaction products.

Upon irradiation, however, the overall loss levels are more erratic than the data for the lower temperature samples. For the samples exposed for a smaller time to this high temperature, the irradiated samples can actually have less conductive loss than the unirradiated samples. This trend has been seen in some of the previous data and may be due to chemical binding of the conducting species. What is more striking is the observation that the growing dipolar loss mechanism, found in the 100°C samples is still present. Therefore, these samples still have a marker for radiation exposure that is visible despite the extreme thermal history of the samples.

Mechanical Data

For all these prototype samples, data on the ultimate tensile strength and the elongation to break were furnished to us by EDF. All the data were taken at a nominal room temperature at a cross head separation rate of 250 mm/min. Data were taken for both the jacket and the central insulation but only the data for the central insulation are reported here, since this is the part looked at dielectrically. The jacket data, however, did show embrittlement.

The data are presented in Table 3 and in Figs. 35 - 38. For all the samples there is a clear increase in the ultimate tensile strength with radiation dose. There seems to be a maximum in the elongation to break for both the 100°C data and the 135°C data with a possible decrease in the 135°C data at 85 Mrad. Both sets of data show that temperature alone does produce some of the same increase, however.

The mechanical data provide evidence for aging. They are not unique enough and do not vary uniformly enough to provide a good measure of lifetime since they do not appear to be sufficiently different for each of the given treatments. If the insulation were to become too brittle, the elongation data would fall to quite small values, as they did for the jacket. From the available data, it is not possible to state as to whether this would occur gradually enough to be useable. In any event, elongation to break is a destructive measure and would require sacrificial samples.

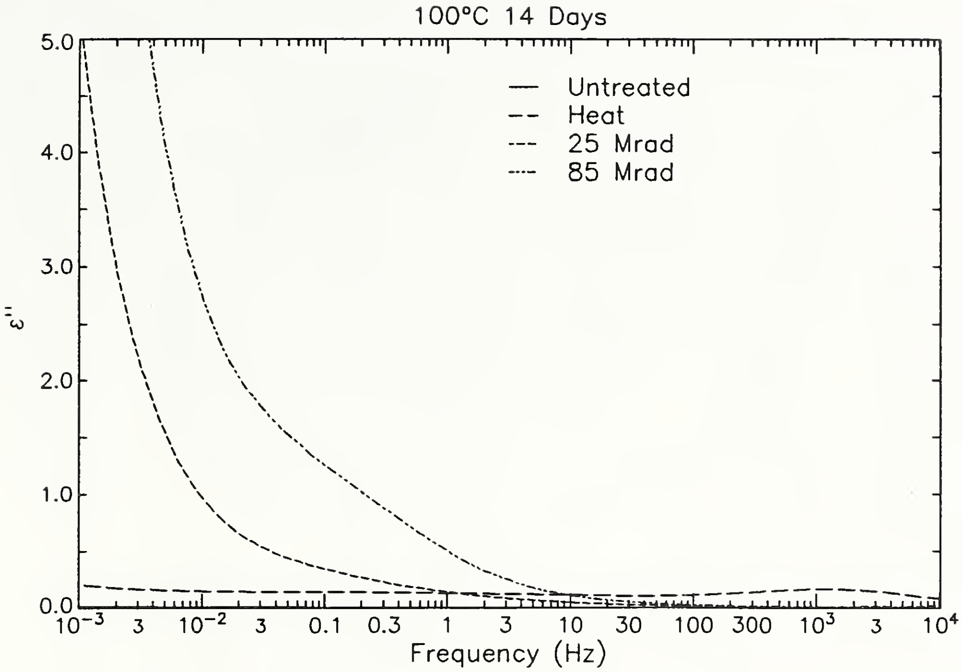

Fig. 23. The loss data for the EDF prototype cable aged at 100°C for 14 days as a function of irradiation. The unaged sample is along the frequency axis.

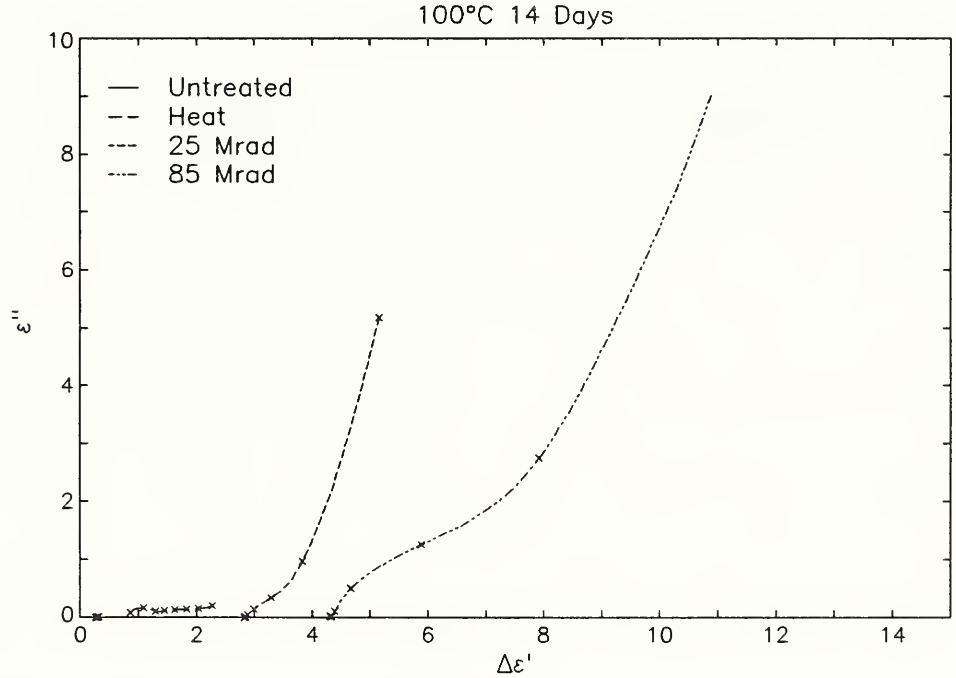

Fig. 24. The loss data for the EDF prototype cable aged at 100°C for 14 days as a function of irradiation replotted in the complex plane. The data for the untreated sample is under the single plotted point.

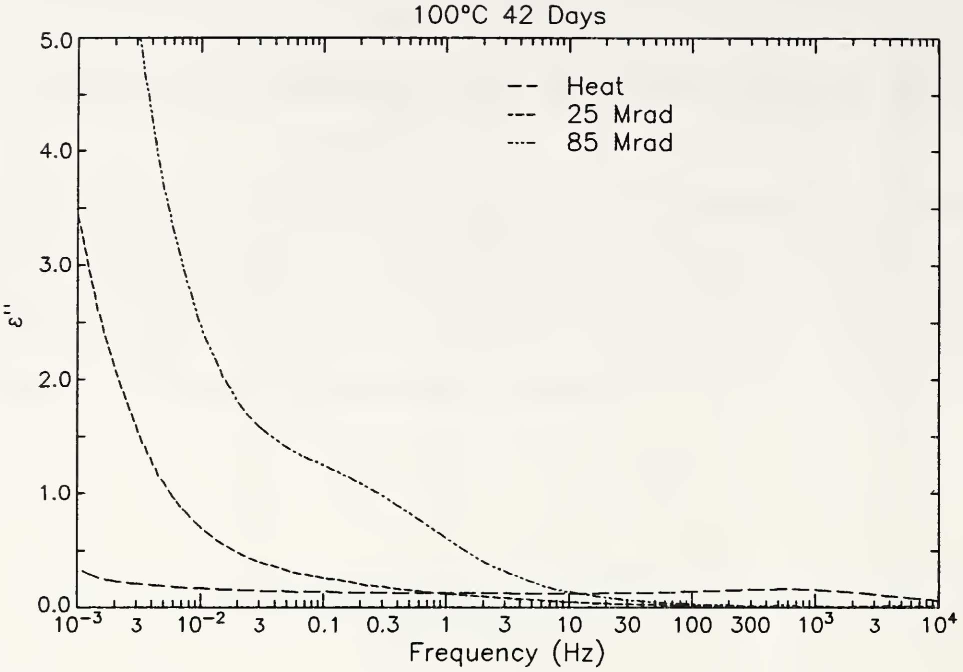

Fig. 25. The loss data for the EDF prototype cable aged at 100°C for 42 days as a function of irradiation.

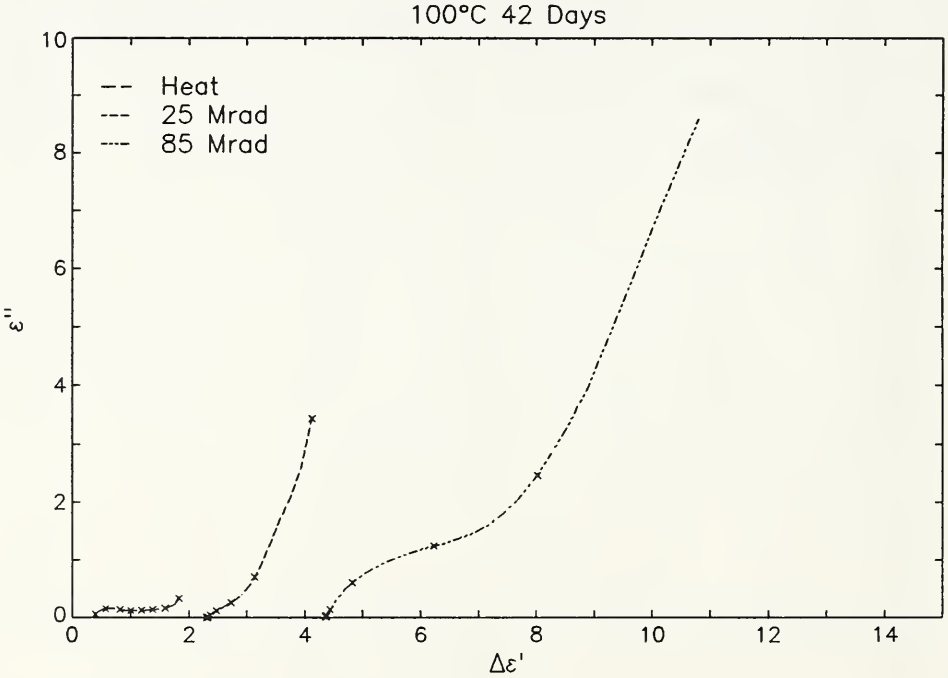

Fig. 26. The loss data for the EDF prototype cable aged at 100°C for 42 days as a function of irradiation replotted in the complex plane.

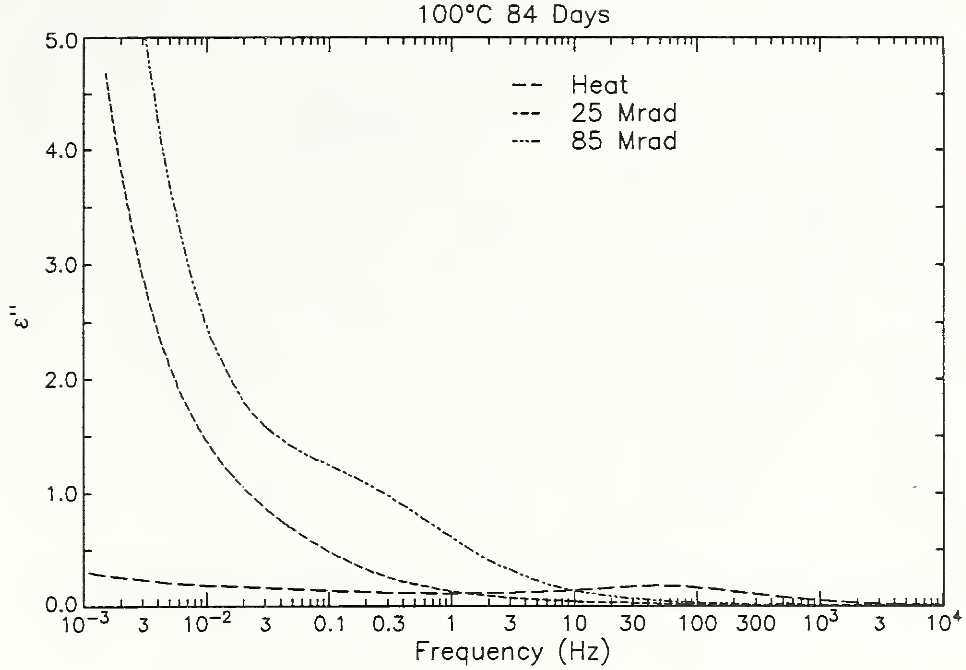

Fig. 27. The loss data for the EDF prototype cable aged at 100°C for 84 days as a function of irradiation.

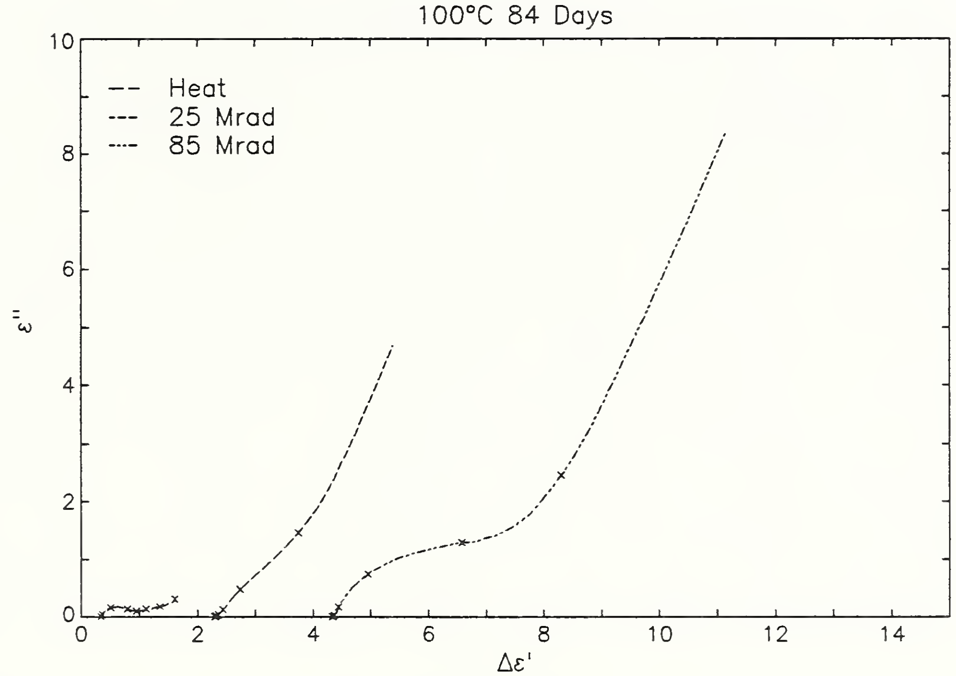

Fig. 28. The loss data for the EDF prototype cable aged at 100°C for 84 days as a function of irradiation replotted in the complex plane.

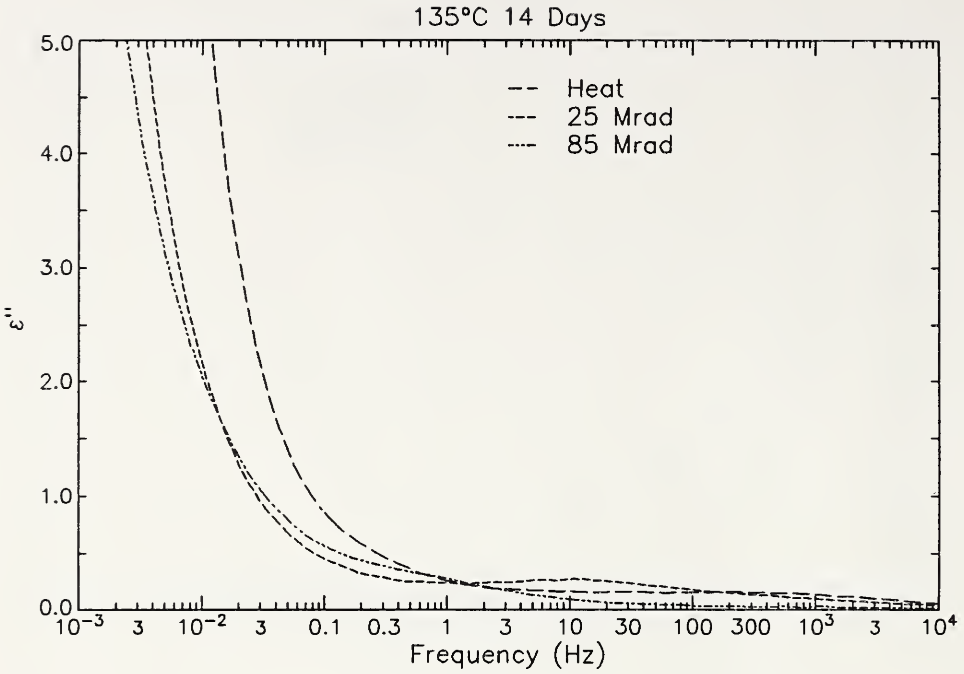

Fig. 29. The loss data for the EDF prototype cable aged at 135°C for 14 days as a function of irradiation.

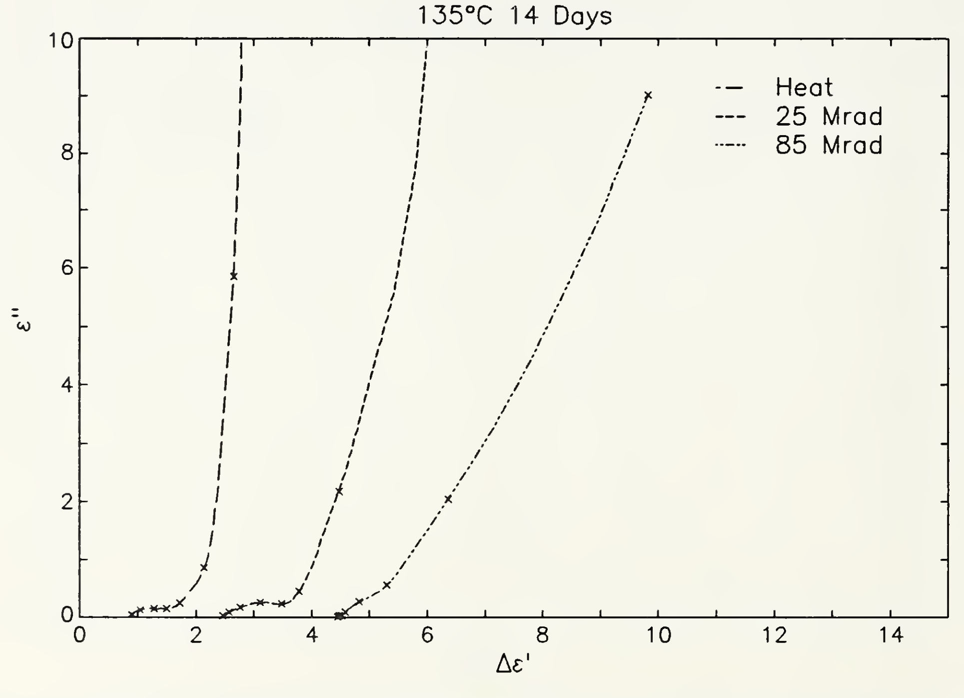

Fig. 30. The loss data for the EDF prototype cable aged at 135°C for 14 days as a function of irradiation replotted in the complex plane.

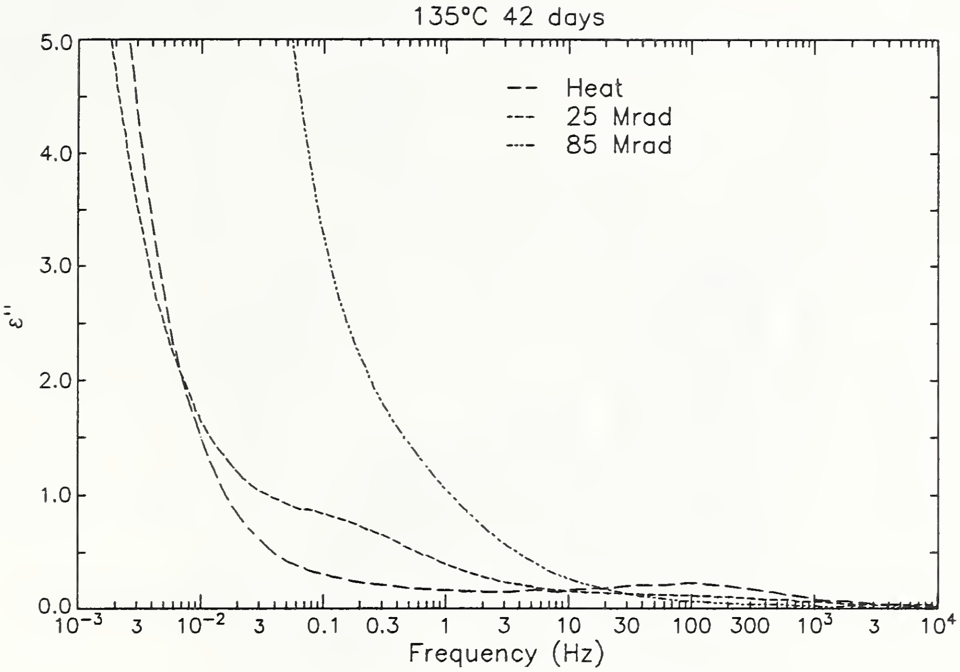

Fig. 31. The loss data for the EDF prototype cable aged at 135°C for 42 days as a function of irradiation.

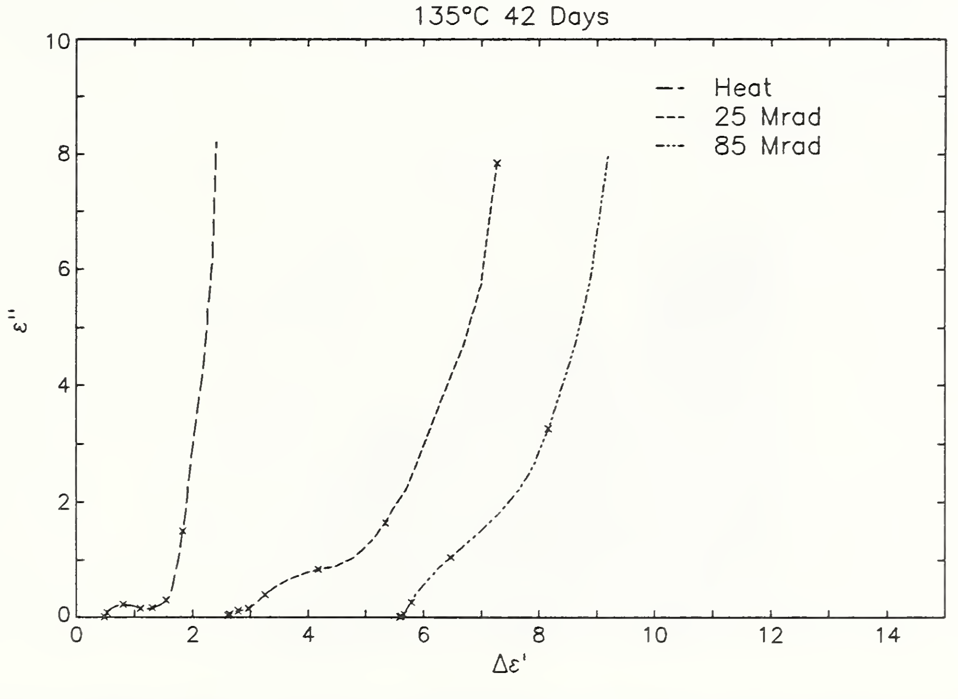

Fig. 32. The loss data for the EDF prototype cable aged at 135°C for 42 days as a function of irradiation replotted in the complex plane.

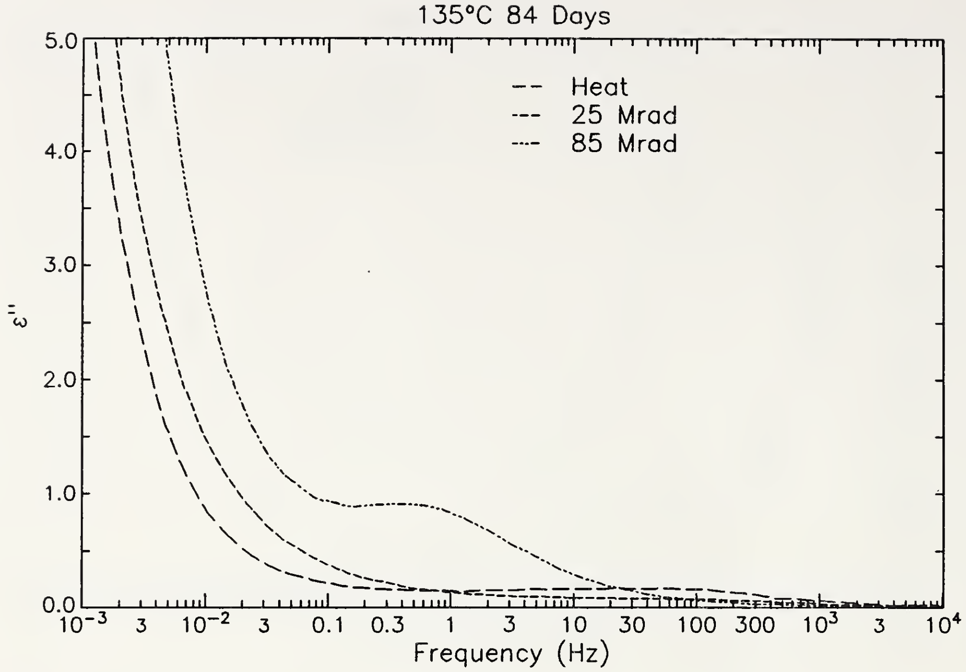

Fig. 33. The loss data for the EDF prototype cable aged at 135°C for 84 days as a function of irradiation.

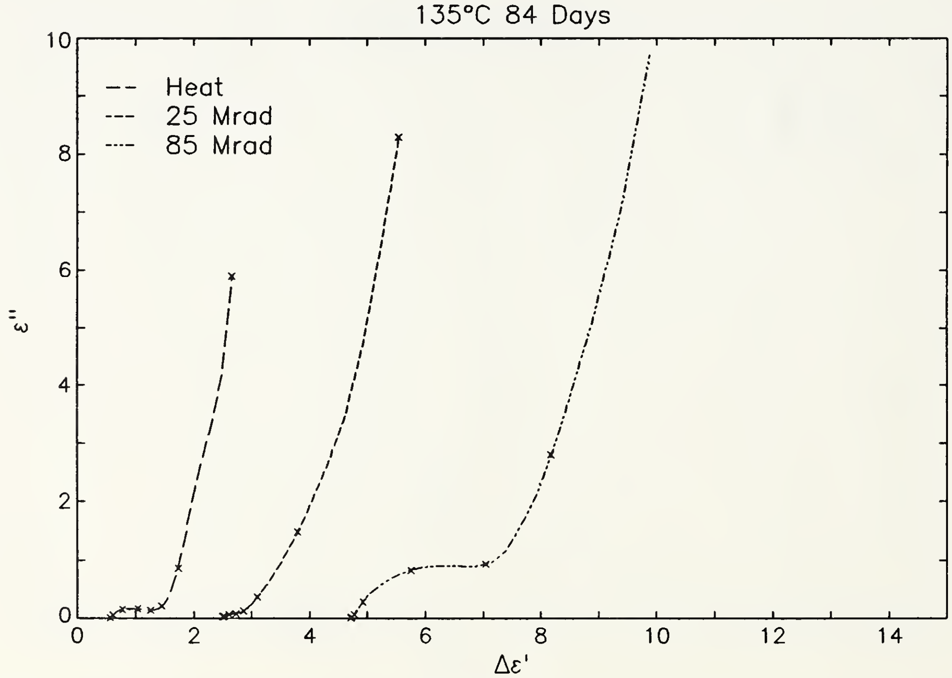

Fig. 34. The loss data for the EDF prototype cable aged at 135°C for 84 days as a function of irradiation replotted in the complex plane.

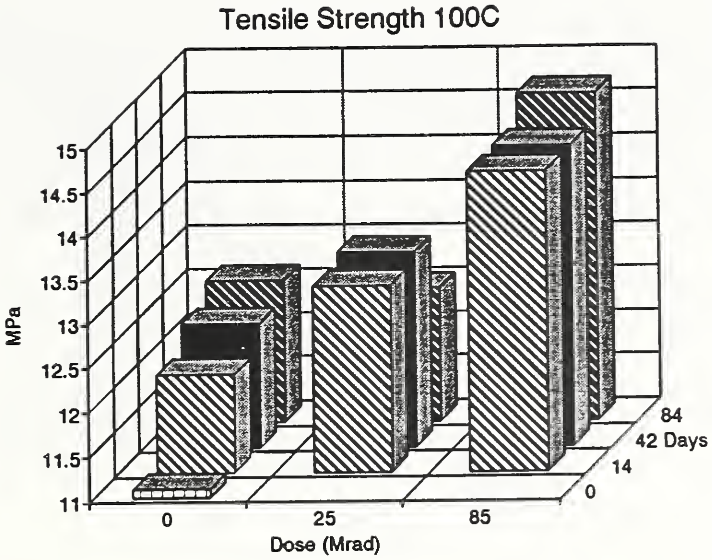

Fig. 35. Ultimate tensile strength of EDF prototype cables aged at 100°C as a function of thermal aging time and exposed dose.

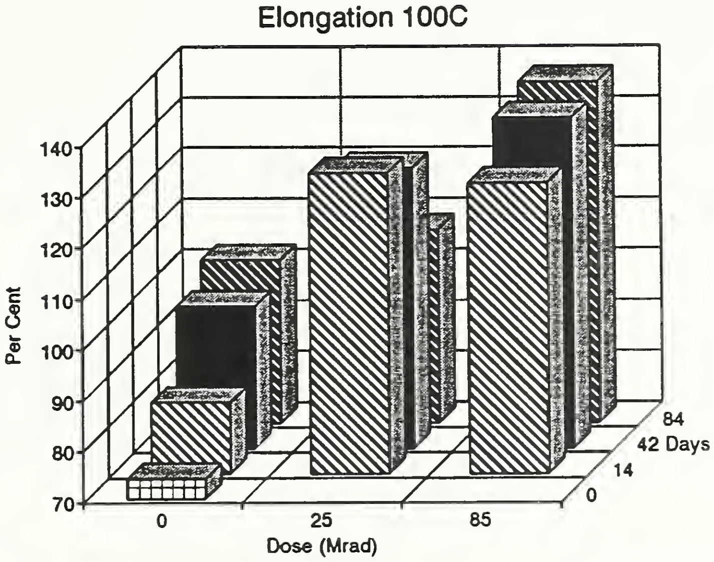

Fig. 36. Elongation to break of EDF prototype cables aged at 100°C as a function of thermal aging time and exposed dose.

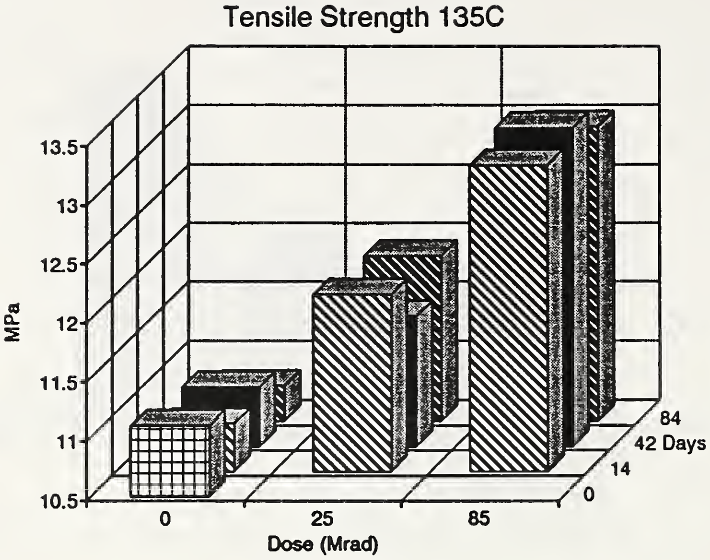

Fig. 37. Ultimate tensile strength of EDF prototype cables aged at 135°C as a function of thermal aging time and exposed dose.

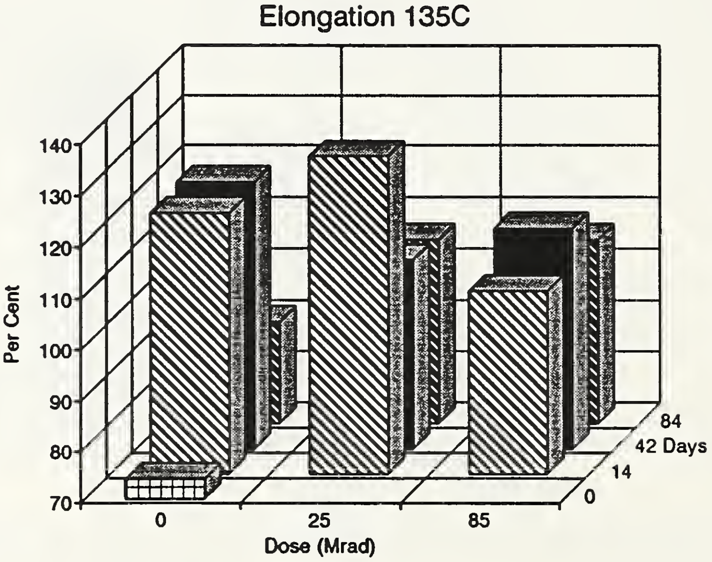

Fig. 38. Elongation to break of EDF prototype cables aged at 135°C as a function of thermal aging time and exposed dose.

DISCUSSION

The aim of the research presented in this report has been to establish the validity of using the Time-Domain Dielectric Spectrometer to estimate the remaining lifetime for cable insulation used inside a nuclear reactor. The background material presented earlier and the data analysis have been included so that the measured results can be gauged as to their significance and importance as a condition monitor. The behavior of any material in a radiation field can be quite complex, and while a detailed description of this behavior is not necessary, a basic knowledge of it can act as a test of relevancy.

For any material test used to follow aging, the property being measured must change significantly in the course of its lifetime. It is the challenge of any test protocol to find such a measure if an early prediction is to be useful. Also, the more directly connected the measurement is with direct consequences of the aging environment, the more reliable the test is likely to be. The results presented in this report on the dielectric measurements of the cables indicate that dielectric constant and loss measurements over a wide, low frequency range do offer this possibility.

Dielectric Measurements

The ability of dielectric measurements to monitor cable aging has been stated previously[16]. The additional data on the EDF prototype cable samples serve to emphasize this point and add a much larger degree of validity to the assertions in that paper. For all the cable dielectric samples examined, there are clear differences that appear upon irradiation. Furthermore, these latest results show that these differences continue to grow with exposure, so that one could possibly estimate the dose that the insulation has experienced.

While no judgment has been made for the end of life from these measurements, as this is outside the scope of this investigation, it would seem that, if a suitable endpoint could be established and measured, it could become possible. What is most important is the establishment of a measurable property that clearly progresses upon aging. While the available samples have not been sufficient to establish any form of aging scale, since only a few had more than one exposure, the current data suggest that it could be achievable. This is especially true if the dipolar loss region could be tied to an aging criterion.

The data also show that the aging rates of a dielectric can be quite sensitive to its formulation. This is apparent in the greatly reduced changes for the EDF Cable 2 where the presence of fire retardant greatly reduced the rate of change of the cable. This reduced rate is in agreement with the change in the rate of cross-linking found previously[7]. If the loss region near 1 Hz proves to be representative of radiation damage proper, then any variations with batch to batch formulations could be revealed by dielectric loss measurements.

The results presented here also show that the hope that a measurable property could be found related to the response of the material is valid. The dipolar loss region near 1 Hz for most of the materials studied in this report, seems to fill the requirement much better than any estimation of conductance. Furthermore, it seems to track the variations in aging for the different insulations studied, but this needs more confirmation.

The present results also show why previous studies on the use of dielectric loss[18] have not been consistent. A single frequency would have trouble distinguishing between the changes in conductance and dipolar loss taking place in a sample since only one point for each frequency spectrum displayed in this report would be available. Also, as most of the data showing aging are below 1 kHz, time-domain techniques such as the Time Domain Dielectric Spectrometer seem to be needed if the required amount of data are to be acquired in a short period of time. For reference, the data presented here required only 12 minutes per determination, exclusive of sample preparation.

These results also show that for different classes of materials, above and beyond normal batch to batch variations, calibration data will be necessary. There are enough differences between different polymers and different formulations that extrapolation could be risky.

Since only one of the samples studied in this report underwent a simulated LOCA and it did not fail, no estimate is possible for knowing how close these samples came to end of life with respect to withstanding a LOCA. The possibility also exists that several different failure modes are possible and their relative importance depends on the use of the cable. Instrumentation cable could fail due to poor electrical properties alone while power cabling might require a short circuit. The corrosion experienced by some of the EDF cables and the breakage of the center conductor suggest yet another possibility. These questions must be resolved for proper lifetime estimation no matter what method is chosen.

Mechanical Measurements

The mechanical measurements in this report are not suitable for a lifetime test as they are destructive tests. They do show that the samples examined in this study have changed mechanically as well as electrically upon aging. They also show that, for the central insulation, it is still quite far from embrittlement. For the EDF prototype cables, at least, any attempt to use an indenter on an intact cable, as has been proposed, would be futile as the outer jacket had become hard and very brittle.

Implementation

The implementation of the Time Domain Spectrometer in nuclear power generating plants has been previously discussed[16]. Just as sample geometries were devised for the test cables examined in this report, similar sampling techniques would have to be implemented for either the actual cables or identical, representative sample pieces. Presumably, the sampling locations would have to be carefully chosen to be those physical locations that subject the cable insulation to the worst possible aging conditions.

Sampling would be important in any application of low frequency dielectric techniques. The measured parameters are the parallel combination of those parts of the insulation between the electrodes. To the extent that the properties vary with position, the average value is measured. Therefore, any sampling electrode configurations would have to be small compared to any spatial variations. Given that the cable pieces examined in this report were 30 cm or less, small size is not a difficult measurement problem.

In any real plant situation, a running reactor could be the source of several interferences. The most obvious one is electrical interference from electrical equipment associated with the reactor. This interference could be minimized by careful shielding of the sample and the leads necessary to connect to outside the reactor. Also, the test could take place at times when no heavy electrical equipment is running.

Another interference, which was not addressed in this report, is from any radiation induced conductivity that decays with time. All the samples used in this study were measured at times long compared to the decay time for such conductivity. This is a problem that would clearly have to be addressed. To the extent that the decay time for such conductivity is not too long, a dielectric measurement could be made when the reactor is idle. Given the fact that cables can survive for tens of years without failure and given that the ideal is a linear scale of aging that progresses well before failure, only periodic measurements would be needed.

Finally, once the end of life criteria are established, the data could be examined for a test method involving just the time-dependent data. With the relevant time scales the order of seconds, proper step response measurements using a measured current might be employed with sufficient resolution.

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Table 1

Dielectric Constant at 10 kHz, 50°C

Cable	$\epsilon_{10\text{ kHz}}$
Shippingport	2.3
Sandia XLPO black	2.6
Sandia XLPO red	2.6
Sandia XLPO white	2.6
Sandia Coaxial	2.2
EDF Cable 1	2.1
EDF Cable 2	2.1
EDF Cable 3	2.2
EDF Cable 4	2.2
EDF Cable 5	2.2
EDF Prototype	2.3

Table 2

Prototype Cable Aging Study

Temperature (°C)	Time (days)	Dose (Mrad)	Condition
none			
100	14	none	
100	14	25	
100	14	85	
100	42	none	
100	42	25	
100	42	85	pale
100	84	none	
100	84	25	pale
100	84	85	pale
135	14	none	light tan
135	14	25	light tan
135	14	85	tan
135	42	none	broken
135	42	25	dark tan
135	42	85	dark tan, broken
135	84	none	brown, broken
135	84	25	brown, broken
135	84	85	black, broken

Table 3

Mechanical properties - Prototype Cables

Temp. (°C)	Time (day)	Dose (Mrad)	Elongation to Break (%)	Tensile Strength (Mpa)
none			74	11.1
100	14	none	84	12.1
100	14	25	129	13.1
100	14	85	127	14.4
100	42	none	98	12.4
100	42	25	125	13.2
100	42	85	135	14.4
100	84	none	102	12.6
100	84	25	108	12.5
100	84	85	137	14.7
135	14	none	121	10.9
135	14	25	132	12.0
135	14	85	106	13.0
135	42	none	122	11.0
135	42	25	107	11.6
135	42	85	113	13.2
135	84	none	90	10.8
135	84	25	106	11.9
135	84	85	106	13.0

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The use of the dielectric properties of insulation to assess the lifetime of electrical insulation in a nuclear power plant is assessed. The factors governing dielectric constant and loss over an extended frequency range are summarized. The Time-Domain Spectrometer is shown to be suitable for the task, given the changes expected in an insulator upon exposure to radiation and the wide range of lower frequencies that it can cover. Several sets of cable samples of the types expected to be found in a reactor were measured with the Time-Domain Dielectric Spectrometer and the results are reported. It is shown that for the hydrocarbon based insulation studied, with the aging conditions used, a characteristic loss region centered near 10 Hz at 50°C is readiation induced and follows dose. This loss seems to be a good marker for radiation exposure and history, even in the presence of large losses due to the presence of filler and other sources of high loss. The use of loss data is proposed as a good possibility for lifetime assessment with further research. Comparison is made with some mechanical measurements that illustrate the enhanced sensitivity of dielectric measurements.

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