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Experimental evaluation and comparison of performance of polymeric insulating materials in radiation environment

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ABSTRACT

Polymeric insulating materials are increasingly being used in outdoor and radiation environments such as space, nuclear power plants etc. It is well known that polymeric materials used for electrical insulations may suffer from tracking. It is necessary to form a credible database about the tracking resistance of different polymeric insulating materials and their behaviour under ac/dc voltage stresses as affected by the radiation. The investigations here show that, the DC resistance to tracking of all the materials investigated were significantly lower than that for AC. It is therefore necessary to formulate separate standards for tracking under DC voltage stresses. Also, the test results show that Silicone rubber and High Density Poly Ethylene samples are reliable for radiation environments.

Introduction

Outdoor insulating bodies have been traditionally made of glass and porcelain materials. These insulators have been tried and tested for many years and have been accepted worldwide. However, polymeric insulators, also called composite or non-ceramic insulators, are becoming popular due to their superior mechanical strength, superior dielectric qualities and better performance in polluted areas. They also have approximately 90% weight reduction compared to porcelain (Gorur et al 1989). So, polymers are currently used as insulation structures in many power system networks. It is well known that polymeric materials used for electrical insulations may suffer from tracking. Tracking, which is a phenomenon of polymeric materials containing carbon atoms in their molecular structure, occurs on the surface because of the creepage discharge resulting from surface contamination. It varies with surface-field intensity, surface-current magnitude and the status of discharge thereby induced, all of which are due to surface wetting and the degree of contamination. There are contaminants that induce tracking such as salt, dust, humidity and atmospheric chemical agents. Once tracking occurs, surface resistive property is lost completely and it never recovers. So, there is a need, to understand the degradation process in the polymeric materials. Artificial ageing techniques may be used to provide a valuable guideline for their design. The artificial ageing of polymers by UV radiation, acid rain, corona discharges are dealt by many researchers.

In recent years with increasing uses of electric and electronic devices in various radioactive environments, which include atomic and nuclear power research, the organic insulation materials are inevitably exposed to various kinds of radiation. Hence, it becomes essential to investigate the influence of such irradiation in insulation materials. The ageing of electrical insulating material has been recognized for many decades as a prime cause of the premature failure of wide variety of nuclear plants (Banford and Fouracre 1999).Nuclear radiation may interact very strongly with materials in general and may cause structural changes that alter their properties. Consequently, it is a topic that has received considerable attention. Much effort has been expended in attempting to understand the processes of ageing and degradation, what types of test should be undertaken to ascertain the condition of insulation and how short term accelerated testing might be carried out in order to forecast the long-term life of insulation systems (Banford and Fouracre 1999).

The items of nuclear power plants that are of particular concern are reactor pressure vessels, its internal components, heat exchangers, piping, pumps, instrumentation and cables. The materials that constitute such items can be subjected to energetic radiation of various degrees as well as other environmental stimuli. From the point of view of nuclear safety the interest in the electrical insulation derives from the fact that they are of vital importance in providing the links between the transducers and the instrumentation and control systems that monitor and control the nuclear power plant. These include the systems necessary for shutting down the plant safely in the event of an emergency.

Ethylene Propylene Diene Monomer, Silicone Rubber, Nitrile Butadiene Rubber and Polyethylene are commonly used in nuclear power plants for producing seals, hoses, electrical insulators and cables (Chipara et al 1999). Insulating materials, when used in such radioactive environment will in normal operation be subjected to a background irradiation throughout its operating life. The irradiation will consist of gamma radiation and neutrons. In an accident situation, the insulation would have to withstand high levels of irradiation. Complete vaporization of the reactor core will be followed by a hydrogen burn of up to 1000˚c. During the period of cool down the insulation will be subjected to condensation of moisture (Banford et al 1999). This suggests that tracking investigations can be carried out on insulating materials used inside the nuclear power plants. This idea is supported by the tracking investigations carried out in polymeric insulating materials used in oil exploration pipes (Venkataraman et al 2005).

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The actual dose rate which the organic insulating materials receive in practice is almost 1 Gy / h in the primary containment vessel of a typical light water reactor (Kuriyama et al 1978, Blodgett and Fisher 1969). Investigations have also been carried out to study the radiation-induced degradation of mechanical properties of electrical insulating materials used in nuclear environments (Chavalior et al 1999, Clavreul 1999, Chipara et al 1999). The radiation-induced degradation in percentage elongation at break of EPDM-NBR rubbers has also been studied at different integral doses between 2 to 200 kGy (Chipara et al 1999). Ageing of Silicone rubber polymers used in electrical equipments in nuclear power plants was studied at different dose rates, with tensile strength as the main parameter for investigation (Clavreul 1999). The authors have investigated the tracking phenomena of SR and EPDM by ageing them under gamma irradiation [24,25]. Herein, a comparison and ranking is made on the performance of the materials aged by gamma irradiation.

According to National Council on Radiation Protection, the average radioactivity per ton of coal used in coal fired power plants is 17000milli curies/4,000,000 tons or 0.00427milli curies/ ton in USA. This figure can be used to calculate the average expected radioactivity release from coal combustion. For 1982, the total release of radioactivity from 154 coal plants in the United States was 2,630,230milli curies. Hence, people living near coal-fired power plants are subjected to more radiation doses than those living near nuclear power plants (Mc Bride 1978 and Gabbard 2005). Hence, it is clear that the outdoor insulators used near coal-fired power plants cannot be dispensed with, the radiation induced degradation, in addition to the other environmental stresses. Hence, researchers in organizations like European Council for Nuclear Research (CERN), Electricite' De France (EDF) and Oak Ridge National Laboratory (ORNL), study the radiation induced degradation of equipments and materials.

Materials used

The commercially available EPDM (EP96), SR (VMQ) and HDPE (EH) supplied by M/S. Rubber Sales Corporation, India were used for the investigations.

Sample Preparation

The samples in the form of sheets were cut into required dimension of 12cm X 5cm X 3mm.The surface of the samples were cleaned using acetone to remove any dust or grease present on the surface and dried.[24,25]

Apparatus and test procedure

The inclined plane test setup used in this work is shown in Figure 1. The test procedure is as per IEC – 60587[10,24,25]. The standard IEC-60587 deals with the test method for evaluation of electrical insulating materials for use under severe ambient conditions at power frequencies (48-62 Hz). The evaluation is done by the measurement of resistance to tracking and erosion, using a liquid contaminant and inclined plane specimens. In this test, the specimen is mounted over a support at an angle 45°. The distance between top and bottom electrodes is 50 mm. The top electrode was connected to HV source and the bottom electrode was connected to ground through 100 Ω resistance to measure tracking current. The potential drop across this was measured using THS-720 P oscilloscope for further analysis of leakage current. To simulate pollution conditions a contaminant solution of 0.1% NH_4Cl is allowed to fall over the specimen at a constant flow rate using a peristaltic pump, Ravel Hitek-RH-P110-S10. Time to tracking is defined as time at which track originating from lower electrode, crosses 2/3rd of the gap between the electrodes or if the complete bulk volume of the material degrades at a particular location due to arcing. Otherwise the test voltage is applied for period of 6 hours and experiment has to be properly terminated.

Ageing of Samples by Gamma Irradiation

The main purpose of this work has been to elucidate the importance of ageing by gamma irradiation in polymeric insulating materials, with respect to long-term performance during use in outdoor in the proximity of nuclear/coal power stations and/or in indoor of the nuclear power plants. With the depletion of the available conventional sources of energy, in future, we may have to majorly resort to nuclear power generations in which case, the study of accelerated ageing of insulators by electro magnetic radiation may serve as a guideline for the material engineer in the process of material selection. Thus, the term performance is understood in terms of both surface electrical properties and resistance to ageing. The surface electrical properties have been characterized by the measurements of tracking resistance, whereas resistances to ageing have been evaluated by various physico-chemical analyses.

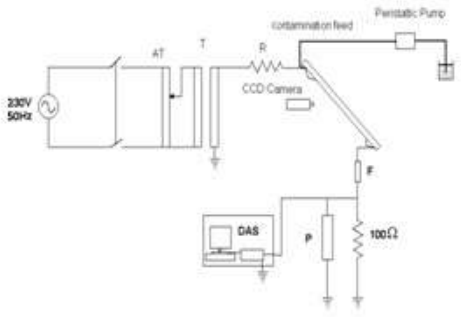

Figure 1. Inclined plane test (AT, Autotransformer; T, HV transformer; 5 kVA, 220V/20 kV; R, Series resistance 33KΩ; P, Protection circuit neon bulb; F, Fuse 100 mA).

The samples of dimension 12cm X 5cm X 3mm were cleaned using acetone and were kept in the the ^{60}Co gamma camber facility, available at the Indira Gandhi Centre for Atomic Research (IGCAR) Laboratory, with an irradiation volume of one litre with a dose rate of 51 kRads/h for the required dose. The time gap between removal of sample from the radiation chamber and mounting it for high voltage test was kept constant (15.30 h) for all the samples. The tracking resistances reported are the mean of five tests conducted at the same test conditions.

Results and discussion

Although considerable work has already been reported in understanding the tracking in AC, it is not possible to realize the basic characteristics of insulation material when it is used for DC application. The DC behaviour of SR and EPDM was studied [24,25]. A comparison of performance of the chosen materials under AC/DC voltage stresses is given in this section. All the samples were tested for their tracking resistances at AC, +ve DC and –ve DC voltages at different conductivities of contaminant. All the tests on virgin samples were repeated five times. To understand the potential safety issues in using the AC tracking test results for DC applications, the applied voltage for DC was kept the same as the AC test. A test voltage of 5kV was maintained. A flow rate of 0.6ml/min was maintained. The conductivity was varied from 1000 to 5000 μS/cm. Note that the recommended 0.1% of NH_4Cl has a conductivity of app 2500 μS/cm. The conductivity range of 1000 to 5000 μS/cm was

selected so as to have conductivity above and below the recommended range.

Samples on ac voltage stress

The tracking variability introduced due to variation in conductivity of the contaminant solution are presented at a constant voltage stress of 5kV AC (rms) for all the chosen materials. Figure 2 shows comparison of tracking resistance of all the materials under AC voltage at different conductivities of the contaminant, at a constant flow rate of 0.6ml/min. All the values indicated are the average of five different tests conducted at the same test conditions.

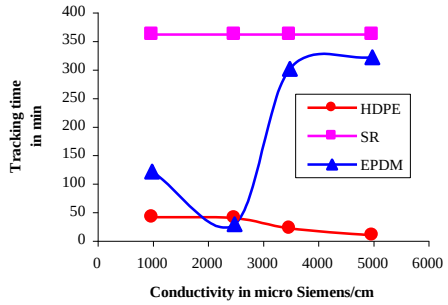

Figure 2.Comparison of performance of materials with the application of AC voltage

It is observed in EPDM that the tracking time decreases with the increase in the conductivity levels. (i.e.) at lower conductivity level, the leakage current through the contaminant raises the surface temperature causing material degradation. With the increase in the conductivity of the contaminant above 2500 µS/cm, the surface discharges are highly arbitrary with a reduced degradation of the material, allowing increased tracking time. The arbitrary discharges reduce the probability of ohmic heating and thereby increase the tracking time[24].

It is observed that in SR, under AC voltages, no samples have failed. This indicates that SR is the ideal insulating material for AC voltages. For HDPE materials, it is observed that the tracking time decreases with the increase in the conductivity levels. The leakage current through the contaminant raises the surface temperature causing material degradation[25].

Samples On +Dc Voltage Stress

As there are no standard tests available for performance evaluation of composite insulators on DC voltages, the results of the performance under AC voltages are generally used in the design of insulators on DC (Du 2001). Hence, to test the possibility of using AC tracking results for the design of DC insulators, a test voltage of 5 kV +ve DC was chosen. Figure 3 shows comparison of tracking resistance of all the materials under + ve DC voltage.

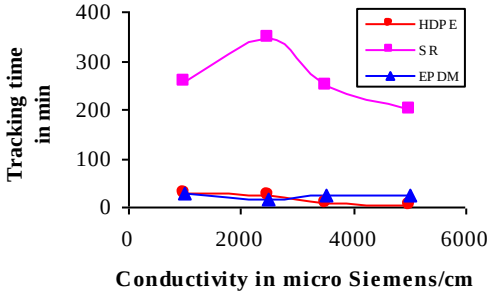

Figure 3. Comparison of performance of materials with the application of +DC voltage

While understanding the tracking results of EPDM material under DC voltages, it is observed that the tracking time of the material under AC voltage is high compared to DC voltages. The results are in agreement with the work done by Du (2001) on polythene films. Similar observations are made for SR and HDPE materials also.

The DC tracking resistance of all the tested material is lower than that of the AC values. This suggests the severity of the electrostatic stress and that of the contaminant on DC voltages. This also indicates the potential safety and reliability issues in power system network if AC resistance to tracking results is applied in the design of DC voltage application. The result of the above investigation confirms that only DC test results are to be used in the design of insulators to be used on DC lines, so that the reliability of the power system is improved. This also necessitates the design of suitable standards for selection of materials for DC insulators [24,25]

Samples on -Dc Voltage Stress

Very few works have been done in understanding the tracking resistance of materials under –veDC voltages. Here, a test voltage of –5kV DC was used, keeping all the other conditions same as the previous two investigations. Figure 4 shows comparison of tracking resistance of all the materials under -ve DC voltage.

It is understood from the investigations that for all the materials, the tracking resistance under –ve DC voltage is less than +ve DC voltage. Hence, it is clear that, insulators with -ve DC voltages are subjected to early failures. Also, the polarity of the DC voltages has to be considered in the insulation design and in the choice of materials.

Ageing Studies

All the samples were subjected to different dose levels of 100, 200, 300 kRads (1, 2 and 3 kGy). The radiation dose of 1 Rad equals 0.01 Gray. All the values of tracking resistance reported are the average of three different tests conducted at the same test conditions.Based on the results obtained on the tracking investigations on irradiated samples of EPDM, SR and HDPE, a comparison is being made to find the suitability of the materials with AC, positive DC and negative DC as applied voltages. Figures 5, 6and 7 show the behavior of different materials under different types voltages.

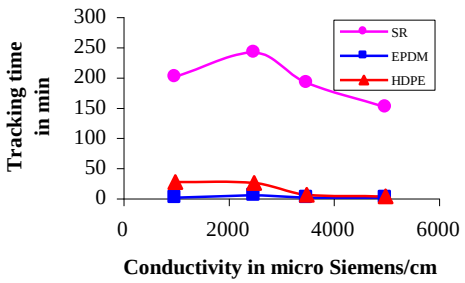

Figure 4. Comparison of performance of materials with the application of -DC voltage

It is observed in EPDM materials that increase in irradiation dose decreases the tracking time under AC voltages for integral doses less than 200kRad. At 200 kRad, this trend changes and this needs to be explained. Under DC voltages of both the polarity, the tracking time was found to be less than that of AC [25].

In SR, ageing by gamma irradiation has no effect under AC voltages. (i.e) All the SR samples withstood the application of AC voltage for 6 hours for all ageing levels. Under DC voltages

of positive polarity, erosions were observed. The tracking time with negative DC voltages were well below that of positive DC. With positive DC voltages, localized erosions were observed than tracking[24].

It is observed that the HDPE material has got excellent tracking resistance under radiation conditions and increase in irradiation dose does not affect the tracking time under AC voltages. This result is in agreement with the tracking investigations made by Du and Kobayashi (2003) on gamma irradiated LDPE material. Under –ve DC voltages, the tracking resistance decreases only slightly.

It is clear that the gamma irradiated HDPE and SR materials withstood the applications of AC voltages. Irradiated HDPE is also found suitable for DC voltages of both polarities. The irradiated Silicone Rubber comes next to HDPE with regard to the performance in DC voltage applications though the performance in negative DC environments is comparatively poor but better than that of the EPDM materials. The photographs taken by a CCD camera during the progress of tracking in EPDM, HDPE and SR are given in figure 8.

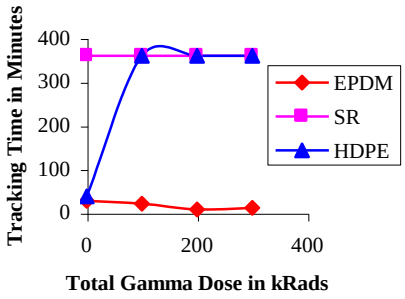

Figure 5. Comparison of performance of aged materials in AC voltage applications

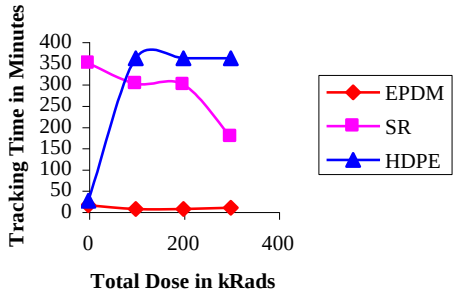

Figure 6. Comparison of performance of aged materials in +DC voltage applications

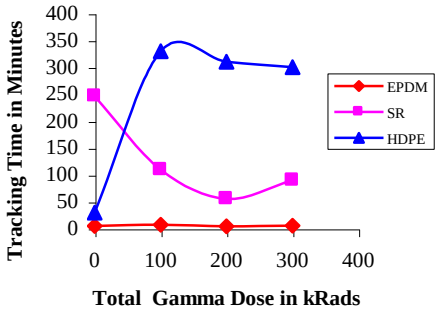

Figure 7. Comparison of performance of aged materials in -DC voltage applications

Physico Chemical Analyses

In order to develop materials with satisfactory resistance to weathering, it is necessary to understand the aging mechanism in a given service environment. The electrical, physical, and chemical properties of the surface of the polymers are critical to the reliable performance of the insulators throughout its service life span. The gamma irradiation may cause damage to the molecular structure of the polymer leading to profound changes in the optical, electrical, or mechanical properties of polymer. This necessitates the use of sophisticated surface analysis techniques such as Fourier Transform Infrared Spectroscopy (FTIR) and Scanning Electron Microscopy (SEM) with Energy Dispersive X-Ray Analysis (EDXA). The effects of gamma irradiation on tracking resistance of LDPE materials were studied by Du and Kobayashi (2003). The reason for the structural modification of LDPE samples aged by gamma irradiation was studied by Banford and Fouracre (1999). It was found that they are suitable for radiation environments and cross-linking is the primary reason for the structural changes of LDPE after irradiation. The results obtained in the present work with HDPE materials agree well with that of the previous researchers. The reason for their good performance was found to be cross-linking reaction, which increases the hydrophobicity of the polymer. Also, Chipara et al (1999) found that thermo plastics respond properly to radiation. The present investigations also prove the same. The contact angle measurements made for aged HDPE samples showed that the hydrophobicity increases after irradiation and this is similar to that observed by Du and Kobayashi (2003). As EPDM and SR have shown degradation in their performances after irradiation, it was necessary to identify the reasons behind their degradation. Hence, only EPDM and SR were considered for all physico-chemical analyses. The virgin samples were used as reference for comparing the surface changes of aged specimens.

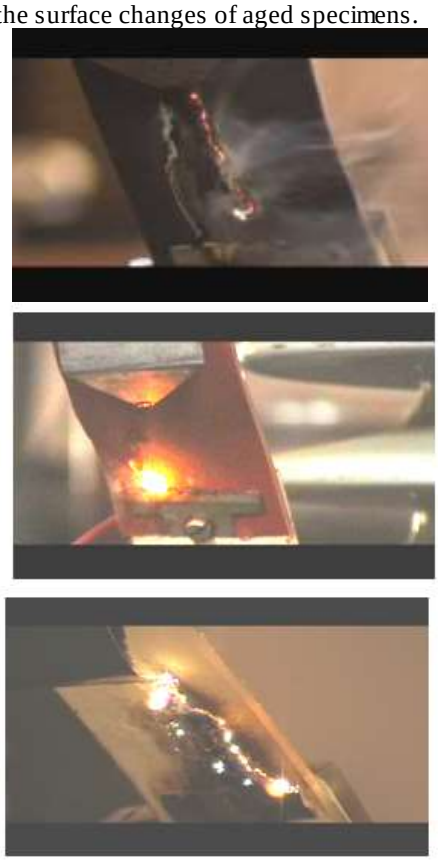

Figure 8. Tracking in EPDM, SR and HDPE

Visual Observations

Visual observations were made on the aged insulating materials. Many polymers discolor during ageing due to oxidation or functional group elimination along the polymer chain (Gorur et al 1989). Severe discolorations were observed in all the irradiated samples of EPDM after subjecting them to high voltage.

These findings are consistent with the previous studies done on polymers aged by different stresses(Gorur et al 1989). A chalky appearance was found around the region of crack for all EPDM samples[25]. This suggests the formation of water-soluble, polar, low-molecular weight degradation products on the surface as suggested by Raji Sundarrajan et al (2004). Surface cracking developed in all the aged samples of EPDM were analyzed. The severity of cracks developed varied significantly.

From the visual inspection of the progressive erosion in SR, the initial degradation was found to be in the form of loss of colour and formation of eroded points near the lower electrode. As time progressed, pitting began and caused damage in depth over the eroded point and the scar of tracking appeared on the edge of electrolyte flowing path. With increase in time, the erosion increased not only in depth but also in length and width. The Figure 9 shows the part of a sample surface of SR failed due to tracking and erosion.

The surface was significantly eroded due to dry band arcing and the eroded surface formed an ash layer with gray colour. The surface near the top electrode was much less affected by dry band arcing.

Figure 9. Sample of SR failed due to a) tracking b) Erosion

Erosion Depth

The maximum erosion depths of tracks were measured using the ULTIMA 200 M2. It is a two channel ultrasonic system at Indira Gandhi Center for Atomic Research at Kalpakkam, which can excite two ultrasonic transducers in a sequence. The data acquired by both the transducers were digitized at a maximum sampling rate of 200 MHz.

For EPDM, the maximum erosion depths were more for samples with –ve DC voltage applications indicating the severity of –ve voltage stresses whereas it was more for aged samples of SR with +ve DC voltage applications as volume erosion occurred for these voltages[24,25].

The Figure 10 shows the maximum erosion depths measured for EPDM with –ve DC voltage, SR aged samples with –ve and +ve DC voltage applications.

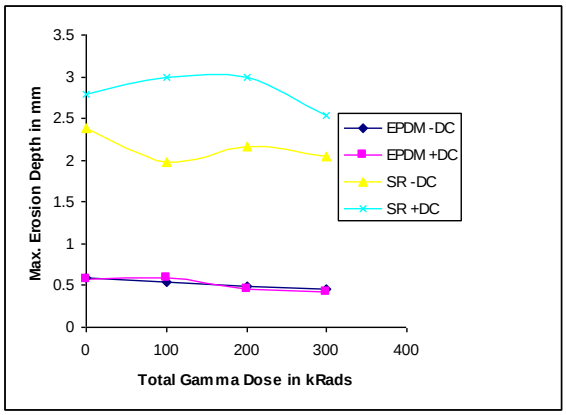

Figure 10. Maximum Erosion Depths of Aged Samples

Contact Angle

Hydrophobicity is the ability of the solid insulator surface to resist the formation of continuous film of water. It is often used to evaluate the ageing process of the surface of the insulating materials. Degradation of the surface of the polymers causes a loss of hydrophobicity, which is usually accelerated with increasing temperature and contamination. The contact angle θ is used to evaluate the hydrophobicity of the surface. Contact angle is a quantitative measure of the wetting of a solid by a liquid. It is defined geometrically as the angle formed by a liquid at the three-phase boundary where a liquid, gas and solid intersect. Low values of θ indicate that the liquid spreads, or wets well, while high values indicate poor wetting. If the angle θ is less than 90 the liquid is said to wet the solid. If it is greater than 90 it is said to be non-wetting. A zero contact angle represents complete wetting.

The contact angle θ was measured on a horizontal surface using Goniometer (ERMA) with a sessile droplet of distilled water after ageing the samples with the required dose of gamma radiation. The conductivity of the water droplet was 2.5 +0.5μS/cm and its volume was about 6.7 μl. The reported measurements of contact angle were averaged over three locations for a given sample. Figure 11 shows the contact angle measurements made for EPDM and SR samples[24,25]. The good performance of polymeric insulating material originates from the fact that polymer surfaces are difficult to wet. However exposure of insulator surfaces to service and environmental stresses changes their chemical composition, modifies their surface morphology and reduces their water repellency.

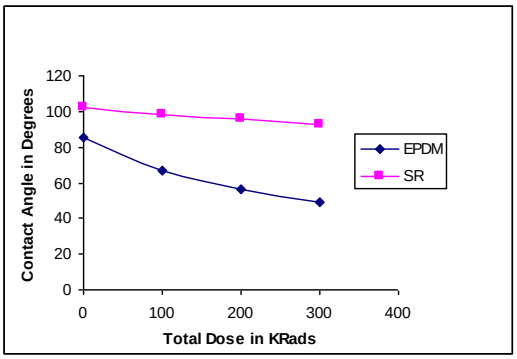

Figure 11. Contact angle measurements on samples

The surfaces of the aged EPDM rubber seem to behave accordingly. This suggests formation of LMW degradation products on the surface of EPDM, which are detrimental to its performance. But, SR surfaces remain hydrophobic even after ageing but the degree of hydrophobicity has come down. The contact angle of the virgin SR sample was 102° and it reduced

after ageing, but only to a smaller extent (93°). Hence, the decrease in the contact angle may be attributed to the early breakdown of the aged EPDM samples.

Scanning Electron Microscopy With Edxa

SEM analyses were performed on all the virgin and irradiated specimens of EPDM and SR, to understand the superficial morphological changes that might have occurred due to ageing. The morphology gives the shape, size and arrangement of the particles making up the object that are lying on the surface of the sample. SEM analyses were performed using a JEOL 840 SEM equipped with an Energy Dispersive X-ray Attachment (EDXA). The size of all samples was 7 mm × 7 mm × 3mm. EDXA was used to determine the elemental composition of the surface.Figure 12 shows the SEM micrographs of virgin EPDM and SR at 25X. The surfaces of both the samples are uniformly smooth and homogeneous and lacked detail.

The manufacturing process tends to bring a layer of the polymer to the surface effectively hiding the filler particles underneath. In Figure 13, the surface of the 200kRad aged EPDM sample where the tracking and the loosely bound filler materials can be clearly seen. It is the SEM micrograph at 500x. The polymer matrix is also degraded.

Figure 12 b shows the SEM micrograph of 200kRad aged SR at 500x. No significant differences between the SEM micrographs of the aged samples and virgin are observed. They show slightly larger number of loose filler particles exposed on the surface of the aged samples.

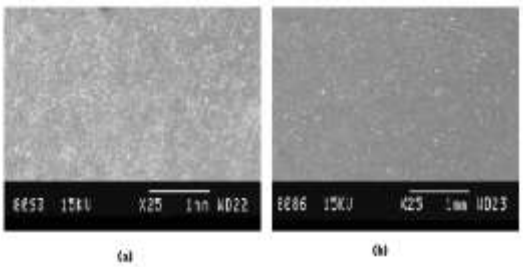

Figure 12. SEM Micrographs of Virgin (a) EPDM and (b) SR

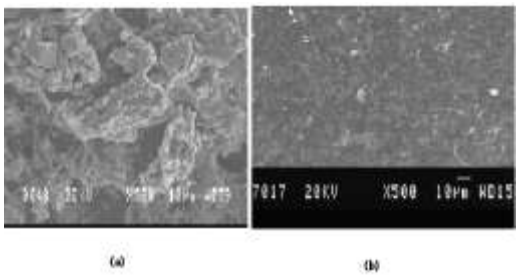

Figure 13. SEM Micrographs of (a) 200kRad aged EPDM and (b) 200kRad aged SR

Energy Dispersive X-Ray Analysis

It uses back-scattered electron imaging. It shows the spatial distribution of elements or compounds within the top micron of the sample. The objective is to determine the possible constituent elements deposited on the surface of the material mainly, the filler concentration because exposition of fillers to the surface of polymer due to chemical changes during ageing are detrimental to the performance of polymers (Raji Sundarrajan et al 2004). Hence, the elemental composition of the surface of the unaged and aged samples of EPDM and SR were determined by EDXA. Figure 14 and 15 show EDXA spectra of virgin, 300kRad aged EPDM and SR samples respectively. The SEM with EDXA reveal that the aged samples of both EPDM and SR show loosely bound filler particles on the surface. silica, S, Ca and Ti were the principal components detected on the surface of EPDM and Si, Ca, Ti were found on the surface of SR.

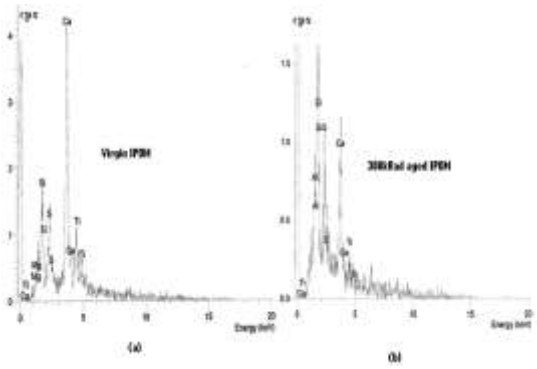

Figure 14. EDXA spectra of a)Virgin EPDM b) 300kRad aged EPDM

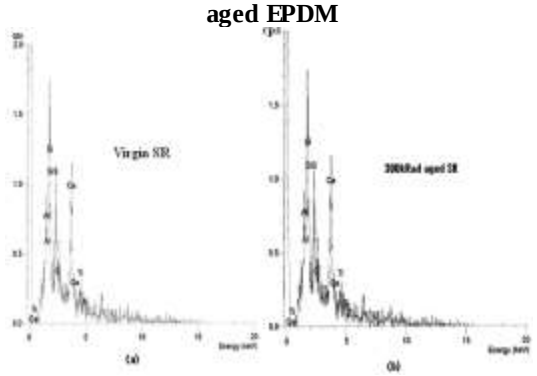

Figure 15. EDXA spectra of a)Virgin SR b) 300kRad aged SR

The EPDM materials consist of C and H as their main backbone elements and a number of fillers such as $Al_2O_33H_2O$, additives, UV stabilizers, antioxidants, etc. The principal components detected were Silica, S, Ca and Ti, the oxides of which are generally added to polymer base to improve the thermal conductivity, electrical conductivity and relative permittivity etc. The EDXA did not detect any Al element, from ATH. Cherney and Gorur also have (1996) observed this. This can be attributed to the fact that Al_2O_3 fillers may be silanized during the manufacturing process or they are so low in quantity. Traces of Si are observed on the surface and are in line with that observed by the previous researchers. They are mainly due to the additives and silicone grease used in manufacturing. Chalking which was mainly observed in EPDM is due to the increase in exposed fillers due to ageing. This is confirmed through EDXA measurements, which showed an increase in Si, Ca and Ti contents. For SR, the increase in surface filler with ageing is negligible for Ca and Ti.

FTIR Analysis

The most important properties of polymers result from their high molecular weights. Their strength results from the entanglement of polymer chains. Degradation of polymers is concerned with the breakdown of macromolecules. This breakdown can be caused by various environmental agencies. Because chemical bonds absorb infrared energy at specific frequencies or wavelength, the basic structure of compounds can be determined by the spectral locations of their IR absorptions. The peak height or the intensity of the absorption bands is proportional to the concentration of the corresponding bonds. Thus, a comparison of the peak heights of IR absorption peaks

will give some insight into the changes in the bonding due to gamma irradiation. The FTIR spectra were taken using Thermo Nicolet Avatar 330 Spectrometer. KBr was used as Beam Splitter. The number of scans is 32 and the resolution of the spectrometer was 4 cm^{-1}. From the visual observations and measurement of erosion depth, maximum damage was found to be for samples aged by 200kRad. So, FTIR spectra for those samples are presented herein in Figure 16.

For EPDM samples, the intensity of the absorption band at 2920 cm^{-1} associated with CH_2 asymmetric stretch had decreased significantly after ageing / degradation. A similar decrease was also observed for the CH_2 absorption band at 2850cm^{-1} associated with symmetric stretch. The next significant observation is the decomposition of methyl CH3 bond with an asymmetric bend at 1425 cm^{-1}. The decrease in the methyl absorption bands had resulted in the loss of hydrophobicity of aged EPDM as observed in Figure 16b. A similar reduction was also observed in CH out-of plane bending at wave number 870 cm^{-1}[25].

The weak bands around 1735 cm^{-1} indicate the presence of carbonyl (C=O) groups formed due to surface oxidation or due to chain scission. The band around 1020 cm^{-1} is related to ATH filler and they show reduction due to ageing. EPDM surfaces react upon exposure to moisture and the alumina filler is reduced to form OH hydroxyl bonds. This is evident from the existence of wide band at wave numbers 3650-3450 cm^{-1}. The sharp bands in these regions indicate that they are intra molecular H bonds. The decrease in these bonds indicates that chain scission had occurred in aged samples. The weak carbonyl absorption groups indicate that surface oxidation is not the dominant reaction due to gamma irradiation.

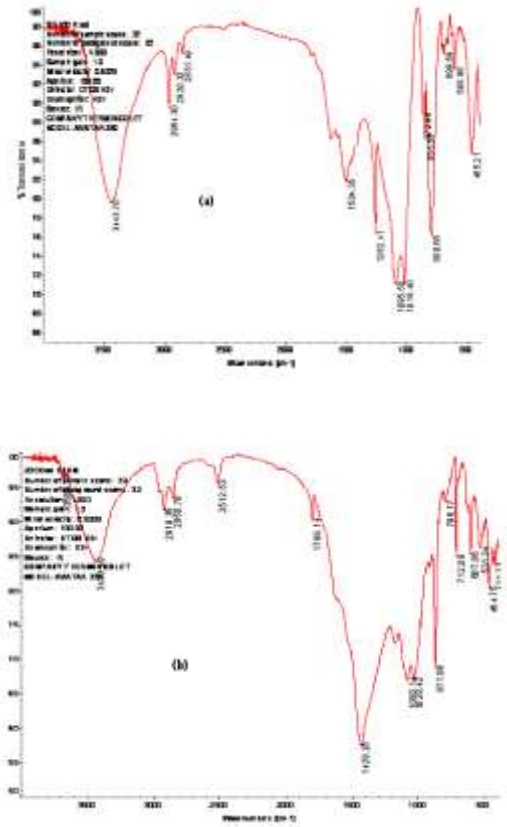

Figure 16. FTIR spectra of a) 100kRad aged b)200kRad aged EPDM samples

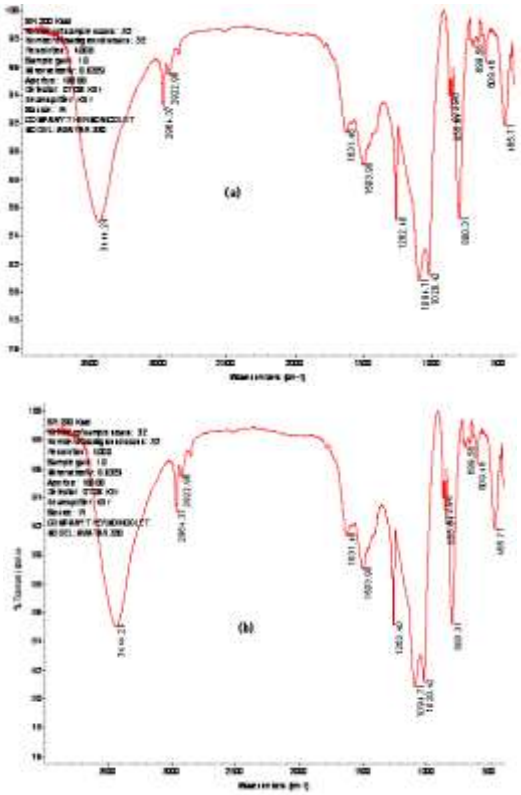

Figure 17. FTIR spectra of a) 100kRad aged b)200kRad aged SR samples

FTIR spectra of 100, 200kRad aged samples are given in Figure 17.Absorption due to OH bonds in ATH at 3400 cm^{-1}, C-H bonds in methyl group at 2960 cm^{-1}, Si-CH_3 bonds (side chain) at 1270 cm^{-1} and Si-O-Si bond at 1100 cm^{-1}appear at the spectra of the virgin and aged samples. From the spectra of the aged samples, the following are observed. There is no trace for carbonyl functional group. This rules out the probability of surface oxidation. The increase in absorption resulting from C-H bonds at 2960 cm^{-1} and Si-CH_3 show net increases in CH_3 functional group in silicone backbone. The Si-O-Si bonds occur in 1000-1130 cm^{-1}. The Si-O-Si absorption bands for stretching vibrations increase with ageing. The presence of silanol (SiOH) stretch at 3400 cm^{-1} shows the reaction of silica to the moisture produced by the electrolyte. The hydroxyl groups bonded with ATH occur in 3300-3650 cm^{-1}. The hydroxyl group formed from oxidation of methyl groups of silicone also gives bands in the region 3200-3700 cm^{-1}. Since, the carbonyl formation is obscured, there is less chance for methyl oxidation.

The aged SR materials show a higher concentration of hydroxyl groups associated with ATH filler. ATH filler decomposes in to alumina and water above 220^{o}C. The fact that the aged samples show lesser surface oxidation suggests that evaporation of water from ATH has occurred. This can be seen from bands around 1630 cm^{-1}. The endothermic reaction of water prevents surface oxidation[24].The bands around 840-790 cm^{-1} are due to Si- alkyl groups $Si(CH_3)_2$. The bands around 870-850 cm^{-1} are due to $Si(CH_3)_3$ functional groups and their changes indicate formation of 2 or 3 dimensional Si-O bonds due to cross polymerization[24].

Conclusion

Based on the results obtained using different evaluation techniques, the following inferences can be made.SR which is

already proved to be the ideal material for AC is found to be the best one for +DC and -DC voltages as well, even under aggravated service conditions. The tracking resistances of all materials under DC voltages are found to be less than that of AC. Very few work has been done in understanding DC resistance to tracking. The results accrued here are consistent with the findings of other researchers who have investigated DC tracking resistances of various other polymeric insulating materials. (Hill 1994, Chang and Mazeika 2000, Sarathi et al 2001, Hirano et al 2001,Uma Maheshwar Rao et al 2002 and Du 2001). The performance of HDPE is inferior to that of EPDM in AC and +DC environments. With –DC voltages, HDPE performs better than EPDM.

It is clear that the gamma irradiated HDPE and SR materials withstood the applications of AC voltages. Irradiated HDPE is also found suitable for DC voltages of both polarities. The irradiated Silicone Rubber comes next to HDPE with regard to the performance in DC voltage applications though the performance in negative DC environments is comparatively poor but better than that of the EPDM materials. Hence, HDPE is found to be the best material for radiation environments within the scope of the doses investigated.

Based on the results accrued from the investigations on the effect of γ - irradiation on polymeric materials, significant changes are found to occur in the performance. a range of measurement techniques has been used to investigate the changes in the behavior of SR and EPDM materials as a result of exposure to γ radiation. The results gave information on the chemical and structural changes within the materials examined. The effect of gamma irradiation and comparison of AC and DC tracking resistances were done by Du and Kobayashi (2003) on LDPE materials and it was found that they are suitable for radiation environments. The results obtained in the present work with HDPE and SR materials also prove the same. Cross-linking was found to be the reason for their good performance.

It is well known that gamma irradiation can produce two distinct effects. The first one involves bond scissions, which in the case of polymers, leads to cross linking and / or molecular weight reduction. The second effect involves the formation of oxygenated compounds, particularly near the surface. it appears that the formation of oxygenated compounds is not significant for both SR and EPDM under the prevailing circumstances. Chain scission was found to be the main cause for degradation in EPDM and cross polymerization was found to occur in SR. The surfaces of the aged EPDM rubber showed loss of hydrophobicity, but SR surfaces remained hydrophobic even after ageing. The fact that the SR and EPDM samples also showed lesser surface oxidation suggests that evaporation of water from the ATH filler protects the polymer. Compared to all the other diagnostic tools used, the FT-IR was found very useful in identifying the chemical changes occurred in the samples. HDPE is recommended to be the suitable indoor insulating material for use inside the nuclear power plants. But, they have to be irradiated before installation. Similarly, the widely used SR insulators are suitable as outdoor insulators in the vicinity of nuclear / coal plants where the background radiation burden is quite significant.

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