Super Electrical Insulation of Polymers in Cryogenic Region

M. Kosaki Gifu National College of Technology Sinsei-Cho, Motosu-Gun, Gifu, 501-0495 Japan E-mail : kosaki@gifu-nct.ac.jp

Abstract: When superconducting power apparatuses and electronic devices are commercially available in the near future, polymers will play a decisive role in the design of electrical insulation. The working temperatures of superconductors regardless of low temperature superconductors at liquid helium temperatures or high temperature superconductors at liquid nitrogen temperature are in the cryogenic region. The polymer insulation at any temperature has a particular role which can not be expected from the gaseous or liquid insulation, namely to support mechanically current carrying conductors. The failure of power apparatuses and electronic devices occur in an insulation part which sustains electric field rather than a conductor part which carries electric current. The performance of the polymer insulation supporting conductors at high potential, therefore, determines the life of every apparatus and device. The capability of electrical insulation of polymers at cryogenic region is generally excellent and it can be named "electrical super insulation" from the criteria of electrical insulation designers at room temperature or above. These aspects will be reviewed in this paper.

DIELECTRIC PROPERTIES

The dielectric constant does not change much and the dielectric loss factor, $\tan \delta$ usually decreases with the decrease of temperature. The thermal degradation and the effect of humidity which affect the dielectric properties at room temperature have negligible effect on insulating materials at cryogenic temperatures. In the genuine solid insulation, it is hardly conceivable to observe any degradation in dielectric properties during or after electric stress application. It is safe to say that if nonpolar polymers such as polyethylene and polypropylene are used, dielectric constant and $\tan \delta$ is low enough to satisfy the cable design. Their $\tan \delta$ in the cryogenic region is in the range of 10^{-5} .[1]

The power loss W due to tan δ is expressed in

$$W = (1/2)\mathbf{w}C\tan\delta V^2 \tag{1}$$

where w is the angular frequency of ac voltage, C the capacitance of the sample and V an applied voltage. The power loss W should be compared with the power loss of superconductors the ac specification and a heat

inleak from environment. It can be said that $\tan \delta$ in the order of 10^{-4} is sufficient to counteract this competition.[2,3] It is obvious that while conductor loss and dielectric loss appear in the ac case, they are zero at dc specification.

ELECTRIC STRENGTH

Polymer Films

When we measure the electric strength E_B of polymer films or more generally film samples, sufficient care has to be taken to obtain the intrinsic data to avoid secondary effects. In particular, this should be noted in the case of measuring E_B of polymer films at liquid helium (LHe) temperature (4.2 K). If one uses liquid LHe as an ambient medium, a streamer of partial discharge in LHe causes a premature breakdown and prevents us from obtaining intrinsic data.

The intrinsic E_B curves in Fig.1 obtained in the recessed sample coated with electrically conducting paint to avoid the effect of ambient media of liquid nitrogen (LN₂) temperature (77 K) [4,5] seems to indicate interesting features. While the nonpolar polymers such as polyethylene and polystyrene have rather flat temperature dependence at cryogenic temperatures, polar polymers such as polyvinyl alcohol or polyvinyl chloride have a monotonic decrease of E_B with temperature. The E_B of nonpolar polymers is explained by the electronic process of breakdown from the thickness dependence at 77 K. Whereas the monotonic decrease of E_{B} of polar polymers with temperature is considered to be related to the polar group in the polymer. However, the steep temperature dependence has not been clarified because the electronic avalanche mechanism does not explain this characteristic.

Composite Materials

Polymers. in general, contract much more than inorganic materials such as metals and ceramics when cooled down to the cryogenic temperature region. In order to adjust the thermal contraction and also reinforce their mechanical strength, glass or plastic fibers and inorganic fillers are incorporated in the polymer matrix. Here we will look at the case of polymers with inorganic fillers.



Figure 1: Temperature dependence of electric strengths of polymers with recessed specimen.

Nonpolar polymers: LDPE (low-density polyethylene); PS (polystyrene); PIB (polyisobutylene). Polar polymers: PVA1 (polyvinyl alcohol); PMMA (polymethyl methacrylate); PVC-Ac (polyvinyl chloride-acetate).

Epoxy and ethylenepropylene rubber (EPR) are commonly filled with inorganic fillers such as silica, alumina, clay and calcium carbonate to add mechanical strength of matrix resin at room temperature or above. They can favorably be applied to cryogenic electrical insulation to adjust the difference of thermal contraction between metals and plastics. Since there are infinite combinations of the kinds of fillers and their amount to fill, mechanical and electrical properties have to be systematically investigated. Fig. 2 shows the result of E_B of EPR as a function of temperature down to LHe temperature.[6] The dc E_B curve shows the characteristics of polar polymer films, namely the increase of E_B with the decrease of temperature. The matrix resin does not have a polar group in its chain.

Therefore the polar group is attributed to the oxidized chain, or the interfacial polarization at the interface between matrix resin and filler particles may be considered. EPR is used in the extruded cable insulation above room temperature. It is, however, far surpassed by crosslinked polyethylene (XLPE) in installations throughout the world because of its inferior electrical insulation capability at the relevant temperature. On the other hand, the excellent mechanical and electrical behaviors of EPR at cryogenic temperatures demonstrate its own potential application to superconducting cable design. The thermal contraction of EPR with fillers is significantly reduced in comparison with pure polyethylene or crosslinked polyethylene as shown in [7].



Figure 2: Temperature dependence of electric strength of EPR film.

DETERIORATION OF POLYMER INSULATION

Partial Discharge

When solid insulating materials are free from defects and have an ideal contact between metal electrodes, partial discharge and subsequent deterioration can be avoided. The 500 kV XLPE cable has been successfully designed and developed for Japanese power industries by elaborate means to eliminate such defects.[8] However, solid insulation might have defects typically shown in Fig. 3 and the gaseous content in the void defect is subjected to the electric field multiplied by the stress enhancement factor to the average field. Because the gas has much a smaller E_B than the surrounding solid partial discharge usually starts in a void defect and degradation prevails.

In the cryogenic temperature region, after cooling from room temperature (RT) the solid material contracts, particularly in organic polymers, and the size of the void defects is squeezed down yielding a favorable situation regarding partial discharge deterioration. Cable insulation with the design of lapping the superconductors with plastic films or papers and immersion with liquid nitrogen or liquid helium has been proposed and practiced.[9] They have excellent behavior in contraction on cooling from room temperature to cryogenic temperatures because the helical lapping structure can absorb the lengthwise contraction in the manner of radial contraction. The drawbacks, however, appear when LN₂ or LHe in the butt gap evaporate, for any reason into GN₂ or GHe. The butt gap becomes the weak spot in the insulation and eventually causes partial discharges. The deterioration of polymer films is severe and, therefore, one has to design the cable insulation in a fairly conservative manner for long term reliability.



Figure 3: (a) Representative defects in practical solid insulator; (b) stress enhancement factors when void defects are present.

Treeing Deterioration

Reasonably thick solid insulation, such as extruded cables, which are free from defects and have a good contact at conductors seldom breaks down instantaneously. Actually, a tree shaped partial breakdown starts from the electrode and grows gradually until ultimate breakdown, typically illustrated in Fig. 4. This phenomenon is called treeing deterioration. There are two types of treeing observed in a conventional extruded cable insulation. One is an electrical treeing and another is a water treeing. The electrical tree starts from the asperities or protrusion on an electrode surface. The water tree starts and grows from the electrode or impurities in the solid insulation where liquid or vapor water and electric field coexist, as shown in Fig. 5.[10] However, water is completely solidified at cryogenic temperatures and, therefore, here the water tree is perfectly excluded. The water tree free condition established is thought to be desirable from the viewpoint of cable designers.

Tree Initiation Voltage

The treeing test is usually performed in the needle to plane electrode system embedded in a polymer block sample. This electrode system introduces non-uniform



Figure 4: Electrical tree through the total cable insulation (by courtesy of Central Research Institute of Electric Power Industry, CRIEPI News no. 97).



Figure 5: Typical pattern of water tree and associated electrical tree.

electric field deliberately and is called a standard treeing sample. This electrode simulates the irregularities on a conductor in real cases. Treeing tests have been tried extensively to establish the long term reliability of organic polymeric insulation. The corresponding treeing tests at cryogenic temperatures started in early 1970 at 77K.[11] The tree initiating ac, impulse and dc voltages (V_{ti}) of a low density polyethylene (LDPE) treeing sample immersed in LN2 are listed in Table 1 along with the results at room temperature. While the ac V_{ti} at 77 K is six times as high as that at room temperature, impulse V_{ti} and dc V_{ti} increase moderately. The very intense electric field at the tip of the needle electrode vielded electro- luminescence which is considered to be due to the alternate injection of carriers corresponding to the ac voltage phase as shown in Fig. 6.[12] Main

poryetnylene.			
Temperature (K)		293	77
Ac V _{ti} (kV rms)		7	42
Impulse V _{ti} (kV)	Positive	+ 25	+ 45
	Negative	- 45	- 70
Dc V _{ti} (kV)	Positive	+ 35	>+ 50
	Negative	- 40	< - 50

Table 1: Tree initiation voltage (V_{ti}) of low density polyethylene.



Figure 6: Electrical luminescence at needle tip in polyethylene (30 kVrms at 77 K).

reason of this increase of V_{ti} is attributed to the high tensile strength of LDPE at 77 K.

The increase of ac voltage V_{ti} in the cryogenic temperature is not limited to LDPE but generally prevails in the other polymers as well, when they are cooled down through their glass transition points, as shown in Table 2.[13] Therefore, it can be said that, generally, the polymeric insulating materials are excellent in terms of treeing resistivity and the exploitation of this characteristic offers incentive to design of superconducting power apparatuses and cable using solid polymers.

Dc Tree and Space Charges

The dc V_{ti} level seems to be much improved at cryogenic temperatures compared to that at room temperature, as shown in Table 1. In the tree initiation mechanism by dc voltage, space charges play a dominant role. The tree initiation by dc ramped voltage increases with decreasing dV/dt magnitude and this result is more prominent when the needle electrode is negative than positive at room temperature or above.[14] This may be explained by the fact that homo-charge injection by electrons from a metal electrode is easier than that by positive holes.

A tree can be initiated when the needle electrode is short-circuited after sufficient time of dc voltage appli-

Table 2: Ac tree initiation voltage of polymers.

	Ac tree initiation voltage (kV rms)		
Temperature	300 K	77 K	
LDPE	7	42	
XLPE	12	35	
EPR	8	29	
PVC (hard)	19.9	21.1	



Figure 7: Initiation ratio in % of short-circuited tree as a function of applied dc voltage; voltage decay time = 150 ns, at room temperature.

cation. This tree is called a short-circuited tree. A large amount of space charge is built up by the application of dc voltage. When the needle electrode is suddenly short-circuited, a high field is produced around the needle tip leading to the formation of a local electron avalanche and tree channel generation. The tree initiation rate of short-circuited tree depends on the time constant of dc voltage decay. The slow decay of dc voltage results in a low rate of tree initiation because it gives ample time for space charges to relax.

Fig. 7. shows the tree initiation rate of a short-circuited tree as a function of dc voltage.[15] The increase in the tree initiation rate with dc voltage is explained by the space charge built-up. The polarity difference is again clear and gives evidence of more pronounced injection of electrons than holes from an electrode. The temperature dependence of the tree initiation rates of a short-circuited tree of EPR, shown in Fig. 8, has interesting features.[16] When the test was thoroughly carried out at 77 K, the tree initiation rate was much lower than the case at room temperature. This may, indicate a smaller amount of space charges accumulated at cryogenic temperatures under the same voltage conditions. This result is confirmed by the experiment where dc voltage is applied to the sample at room temperature and it is subsequently cooled down and short-circuited at 77 K. Space charge injection is easier at room tem



Figure 8: Temperature dependence of initiation ratio of short-circuited tree; voltage decay time = 150 ns.

perature than at cryogenic temperatures.[16] Considering these results along with the enhancement of E_B of polymers at cryogenic temperatures, one is tempted to design the electrical insulation of a dc superconducting cable using extruded polymers.

SUPER ELECTRICAL INSULATION

The electrical insulation properties of mainly polymeric materials at cryogenic temperatures have been reviewed. Their extraordinary characteristics as compared to those at room temperature or above can be summarized as follows.

- 1. Low dissipation factor or tanδ
- 2. High electric strength
- 3. Extremely high treeing resistivity
- 4. Water tree free
- 5. Partial discharge free if voids are totally eliminated
- 6. Thermal or chemical deterioration free

They are apparently ideal material characteristics when it comes to design electrical insulation of power apparatus and cables. In other words they can be called "super electrical insulation". However, one should not forget the drawbacks of polymeric materials at cryogenic temperatures. The first is the large thermal contraction when they are cooled from room temperature down to cryogenic temperatures and the second is their brittleness in the cold state. If these drawbacks are overcome, it is possible to exploit the best of their characteristics. The authors have tried a successful method for this subject. That is, a combination of polymers with appropriate amounts of inorganic fillers. The variety of the filled polymers is abundant in terms of kind of fillers and their mixing amount.

Typical filled polymer, EPR was employed for the electrical insulation design of extruded polymer insulated superconducting cable incorporating niobium superconductors operating at LHe temperature.[17] The ac superconducting cables with relatively conservative specifications of 20 kV, 20 kA and 15 m in length, equipped with two terminal bushings also having EPR insulation, were designed and developed by the authors as shown in Fig. 9. They were successfully cooled down to LHe and energized simultaneously by the voltage and current application. The authors also tried the short EPR insulated high temperature superconducting model cable with bismuth type high temperature superconductors cooled to LN2 temperatures.[18] These efforts proved that the exploitation of super electrical insulation capability of polymers at cryogenic temperatures may be applied at various phases of insulation design in the superconducting and cryogenic electrical devices.

CONCLUSIONS

The electrical insulation properties of organic polymers and their composite with fillers in the cryogenic temperature region are reviewed.

 The dielectric properties are satisfactory and less problematic. The dissipation factor, tan
decreases with the decrease of temperature and usually creates negligible or tolerable heat generation compared to the ac loss of conductors and the heat inleak from the environment.



Figure 9: Extruded EPR-insulated superconducting cable.

- 2. The electric strength (E_B) of non-polar polymers is temperature independent and is much larger at cryogenic temperatures than at room temperature (RT). On the other hand, E_B of polar polymers decreases with temperature and polyvinyl alcohol gives 15 MV/cm at 77 K. Generally speaking E_B of polymers are significantly higher than those at RT.
- 3. Plastics with fillers, in the case of ethylene propylene rubber (EPR), give excellent electrical performance in addition to the controlled thermal contraction at cryogenic temperatures.
- 4. Partial discharge deterioration need not be considered if the solid is free from defects, because polymers contracts heavily and inadvertently introduced voids are squeezed out. However, if partial discharges occur in the butt gap at cryogenic temperatures, deterioration may proceed more malignantly than in the case encountered at room temperature.
- 5. Ac tree initiation voltage (V_{ti}) of low density polyethylene at 77 K is six times higher than V_{ti} at RT. The higher tensile strength and E_B and prohibition of auto-oxidation can explain the higher treeing resistivity. On the other hand, dc and impulse V_{ti} are moderately improved in the cryogenic temperature region because those V_{ti} at RT are usually very high. This improvement in treeing resistivity can be expected in almost all polymers if their glass transition point is lower than RT.
- 6. Dc short-circuited tree characteristics are also significantly improved at cryogenic temperatures, partly because the space charges injected from a needle electrode are limited compared to that at RT.
- 7. Plastics at cryogenic temperatures are bound to have desirable insulation capabilities as mentioned above which can be called "super electrical insulation". If the thermal contraction of plastics can be controlled by the amount of fillers, a dependable insulation design is expected. Research and development were carried out for the extruded EPR insulated low temperature ac superconducting cable. It was cooled to 4.2 K and performed well with the simultaneous voltage and current test. The improved dc short-circuited tree resistivity at cryogenic temperatures may open an avenue to filled plastic insulated dc high temperature superconducting cables.

REFERENCES

[1] Chants, M.J., "Dielectric properties of some insulating materials over the temperature range 4.2 ~ 300 K", *Cryogenics*, vol.7, p.351, 1967.

[2] Kosaki, M., Nagao, M., Mizuno. Y., Shimizu. N. and Horii, K., "Development of extruded polymer insulated superconducting cable," *Cryogenics*, vol.32, pp.885-894, 1992.

[3] Forsyth, E.B., "Energy loss mechanism of superconductors used in alternating-current power transmission system," *Science*, vol.242, pp.391-399, 1988. [4] Oaks, W.G., "The intrinsic electric strength of polyethylene and its variation with temperature," Proc. IEE, vol.95, pp.36-44, 1948.

[5] Ball, I.D.E., "The intrinsic electric strength of polyvinyl alcohol and its temperature variation," Proc. IEE, vol.98, pp.84-89, 1951.

[6] Mizuno. Y., Nagao, M., Kosaki, M., Shimizu, N. and Horii, K., "Evaluation of ethylene-propylene rubber as electrical insulating material for a superconducting cable," IEEE Trans. Electr. Insul., vol.27, pp.1108-1117, 1992.

[7] Mizuno, Y., Nagao, M., Kosaki, M., Shimizu, N. and Horii, K., "Development and tests of extruded polymer insulated superconducting cable," 6th International Symposium on High Voltage Engineering, 13.24, pp. 1-4, 1989.

[8] Kubota, T., Takahashi, Y., Sakuma, S., Watanabe, M., Kanaoka, M. and Yamanouchi, H., "Development of 500 kV XLPE cables and accessories for long distance underground transmission line-Part I: insulation design of cables," IEEE Trans. Power Deliv., vol.9, pp.1741-1749, 1994.

[9] Forsyth, E.B. and Thomas, R.A., "Performances and summary of the Brookhaven superconducting power transmission system," *Cryogenics*, vol.26, pp.599-614, 1986.

[10] Shaw, M.T. and Shaw, S.H., "Water treeing in solid dielectrics," IEEE Trans. Electr. Insul., vol.19, pp.419-452, 1984.

[11] Kosaki, M., Shimizu. N. and Horii, K., "Treeing of polyethylene at 77K," IEEE Trans. Electr. Insul., vol.12, p.4045, 1977.

[12] Shimizu, N., Kosaki, M. and Horii, K., "Space charge effect on local electric breakdown of polyethylene at 77K," *J. Appl. Phys.*, vol.48, pp.2191-2195, 1977.

[13] Shimizu, N. and Tateno, Y., "Effect of degassing and low temperature on electrical treeing in ethylene based copolymers," IEEE Annual Report Conference on Electrical Insulation and Dielectric Phenomena, pp.695-700, 1993.

[14] Nawata, M., Kawamura, H. and Ieda. M., "Treeing breakdown phenomenon in polyethylene by dc voltage (in Japanese)," Trans. IEE Japan, vol.95A, pp.423-430, 1975.

[15] Srinivas, M.B. Minoda, A., Nagao, M. and Kosaki, M., "Electrical treeing in EPR at room and cryogenic temperature and its fractal characteristics," 5th International Conference on Conductance and Breakdown in Solid Dielectrics, pp.309-313, 1995.

[16] Minoda, A., Mizuno, Y., Nagao, M. and Kosaki, M., "Dc short-circuit tree in EPR at cryogenic temperature," International Symposium on Electrical Insulation Materials, IEE, Japan, p.32, 1995.

[17] Kosaki, M., Nagao, M., Minoda, A., Mizuno. Y., Hirata, N., Nagata, M. and Tanaka, S., "Development and tests of extruded ethylenepropylene rubber insulated superconducting cable," *Cryogenics*, vol.35, pp.805-807, 1995.

[18] Kosaki, M., Minoda, A. and Nagao. M., "Feasibility study of extruded polymer insulated dc superconducting cable," 10th International Symposium on High Voltage Engineering, vol.4, pp.117-120, 1997.