

# Insulating Materials at Very Low Temperatures: A Short Review

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**Abstract**—In this paper, a short review is given on insulating materials at very low temperatures. Various insulating materials are investigated in terms of phenomena such as partial discharges. Some of the factors affecting the behavior of the insulating materials at very low temperatures, such as the quality of the electrode surface, the stressed insulation volume, and the existing bubbles, are also reported and commented upon. Proposals for future research are also discussed.

**Keywords**—superconductors; high temperature superconductors (HTS); cryogenic insulation; liquid nitrogen; partial discharges; streamers; aging of materials

## I. INTRODUCTION

Since the discovery of superconduction in 1911, there were plans for the design of superconducting cables [1]. After several decades from this discovery, the development of superconducting cables seemed to be possible. Since the mid-eighties of the last century, there is a drastic development in superconducting research because of the discovery of high temperature superconductors (HTS) in the critical temperature  $T_c > T$  of liquid nitrogen (77K) by Georg Bednorz and Karl Mueller of IBM, Rueschlikon, Switzerland. Let us notice that the power needed at room temperature for the function of a refrigerator at 77K is lower than the one tenth of the power needed for the function of a refrigerator at 4 K. Regarding superconductivity and its possible applications, it becomes apparent that electrical insulation is pivotal to the development of reliable energy systems. A breakdown of the electrical insulation may have catastrophic consequences for a superconducting energy system. A HTS superconducting cable offers reduced losses, it is more environment-friendly, it increases the reliability of the system and, it is more flexible than most traditional cables. There is a hope that HTS superconducting cables may give the solution for large urban centers in the future, as the global population increases and the demand for electricity becomes more acute. To be sure, the new technology has to prove that its reliability is at least as good – if not superior - as that of the conventional systems. Such superconducting cables must withstand probable pressure variations as well as some electrical overstressing. In the context of this paper, some aspects of the electrical insulation for superconducting applications will be mentioned and

analyzed. Moreover, some proposals for future research will be made.

## II. SOME GENERAL COMMENTS

Electrical insulation is crucial to the good performance of very low temperature energy systems or for any energy system for that matter. Typical applications of superconducting systems are the superconducting faults current limiter (SFCL) and the superconducting magnetic energy storage system (SMES). HTS transformers are also a possible alternative to conventional transformers since they reduce the losses by 60% or even more. They are more environment-friendly and they use liquid nitrogen instead of mineral oil as insulation and coolant. They are more reliable than the conventional transformers. Their possible applicability has been demonstrated in countries such as Japan, the USA, Germany, South Korea and New Zealand [3]. HTS cables have been built in the past few decades. Such cables use as insulation either non-polar polymer films with liquid helium or nitrogen, or polymer films such as cross-linked polyethylene XLPE. In the case of underground cables, insulating materials that have been used for conventional applications, have been adapted to HTS applications. Consequently, materials such as polypropylene laminated paper (PPLP) [4], ethylene propylene rubber (EPR), XLPE, low-density polyethylene (LDPE) and PTFE have been investigated accordingly [4-6]. However, with the requirements for higher voltages (e.g. 275kV), superconducting cables had considerable dielectric losses [7]. In such voltages, in order to reduce dielectric losses, other insulating materials were used (e.g. Tyvek – a polyethylene nonwoven fabric – as opposed to conventional PPLP, or the ultra high molecular weight polyethylene (UHMWPE)) [8-10].

## III. ON SUPERCONDUCTIVITY

Superconductors transfer practically current with zero losses when their temperature falls below a certain value, which is called “critical temperature”. Superconductors preserve their properties in a region which is defined by three quantities, namely the critical current density  $J_c$ , the critical temperature  $T_c$  (the temperature at which the resistance becomes zero) and the critical magnetic field  $H_c$  (as the magnetic field increases the superconductivity of the material is

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reduced). The higher the values of all three aforementioned quantities, the easier becomes the application of the superconductors. Increase of one of the aforementioned quantities may return the material to its previous (non-superconducting) state. When a superconductor loses its superconducting state, it needs a considerable amount of time in order to revert to superconduction, whereas when a superconductor has an excessive current, the consequences may be deleterious because of the released heat [11, 12]. HTS superconductors are more efficient than common superconductors which function at lower critical temperature. HTS superconductors are in superconducting state at temperatures higher than 77K in applications with weak magnetic fields. On the contrary, materials of lower critical temperatures, such as Nb-Ti and Nb<sub>3</sub>Sn, must be cooled at 4K in order to have superconducting properties. However, in this way, their cost rises [13].

Regarding high voltage cables, there are two main categories of cables, the cables of warm dielectric (WD) and the cables of cold dielectric (CD). Cables of WD have liquid nitrogen whereas the solid insulation can be at room temperature. Their insulation can be XLPE, PPLP or ethylene propylene rubber (EPR). The insulation is applied on the outer wall of the cryostat. This type of cables presents higher losses, which in turn cause higher released amounts of heat [14]. In the CD cables, the insulating material is on the inner wall of the cryostat. In this case, the liquid nitrogen functions both as coolant and as dielectric. This type of cables has an increased cost but, on the other hand, has lower losses and a higher conduction in higher currents [13, 15]. In the Triax cables, all three phases are in a single cable. Such a construction reduces the electric losses and requires half of the superconducting material compared with the CD cables. Triax cables occupy less space than the aforementioned WD and CD cables [13].

#### IV. INSULATING MATERIALS FOR SUPERCONDUCTING APPLICATIONS, PARTIAL DISCHARGES, AND ELECTRICAL TREES

As far as cables at very low temperatures are concerned, there are four types of insulations: 1) vacuum insulation with solid insulators, 2) liquid insulation with solid insulators, 3) tape insulation impregnated with liquid helium or liquid nitrogen, and 4) liquid helium with solid insulation [4, 16]. It goes without saying that in the above cases, the solid insulation must be compatible with the liquid or the vacuum, that the thermal expansion or contraction must be within certain limits and that the cost of construction must be reasonable [17]. Generally speaking, an insulation is affected by factors which tend to reduce its molecular structure, such as high temperatures (which influence the conductivity and the generation of partial discharges), mechanical stressing (which reduces the distance between the electrodes), affecting thus the breakdown strength [18]. Partial discharges are very dangerous in very low temperatures since they produce significant amounts of heat, leading in this way to the total failure of the insulation [1, 16]. In the case of superconducting insulation, Partial Discharges (PD) may contribute to the increase of dielectric losses. A way to reduce such effects is to increase the pressure, preventing therefore the creation of bubbles. In this way, PD activity is reduced and the inception voltage increases.

PD measurements are considered crucial for cryogenic applications because PD are the main source of aging and breakdown of insulations in such low temperatures. Various techniques have been proposed in relation to such measurements. Phase-Resolved Partial Discharge (PRPD) measurements as well as the Current Pulse Waveform Analysis (CPWA) measurements have been proposed for a consequent study of PD in very low temperatures insulating systems. Whereas the first has the advantage of offering an overall picture of the state of the insulation, the latter method can obtain detailed individual PD currents pulses together with the time transition from PD inception to breakdown with the PD detection sensitivity of 0.1pC [4, 19–22]. It has to be noted that during the past few years, a new technique was developed [23]. The latter technique is named Chaotic Analysis of PD (CAPD) and is based on three normalized parameters obtained from two consecutive PD pulses, i.e. amplitude difference ( $P_t$ ), occurring time difference ( $T_t$ ) and correlation between  $P_t$  and  $T_t$ . Authors in [23] claim that besides PRPD, their proposed method is suitable for HTS applications.

Electrical treeing in polymers at cryogenic temperatures may also be noted. Electrical trees may be the result of PD and/or some mechanical stressing. Inception voltages for treeing are in general higher than the inception voltages at room temperature, and even if the electrical trees start, their development is much slower [17, 24]. In [24], a significant point is made, namely that, if a polymer is defect-free, it contracts heavily at very low temperatures and - inadvertently introduced - voids are squeezed out. However, if PD occur in a void at cryogenic temperatures, deterioration may proceed more rapidly and malignantly than in the case encountered at room temperature.

#### V. HTS INSULATION

Insulation is crucial to the successful performance of superconducting apparatus [25]. The characteristics of liquid nitrogen (LN<sub>2</sub>) are fundamental to many HTS applications. Moreover, the role of composite insulating systems (from LN<sub>2</sub> and solid insulating materials, such as paper and epoxy) is also very important, as well as the gaseous nitrogen (GN<sub>2</sub>) and the vacuum [21]. For the correct, reliable and practical design of an HTS insulation is necessary to examine factors that are crucial for the cryogenic liquids, such as the state of the electrode surfaces, and their effect on the breakdown strength [26]. As critical factors influencing the PD behavior and the breakdown strength of LN<sub>2</sub> are the quality of electrode surfaces, the volume of insulation, the presence of bubbles and foreign matter [27, 28].

#### VI. INFLUENCE OF ELECTRODE SURFACE, STRESSED VOLUME AND BUBBLES

It is well known that the breakdown strength of conventional insulating materials is affected by the electrode surface (quality of the surface as well as size of the electrodes) and the stressed volume [29, 30]. Relatively recently, it was shown that the breakdown strength decreases with the increase of the gap spacing and the electrode area for sub-cooled nitrogen (SLN<sub>2</sub>). Such phenomena were observed because the amount of impurities and bubbles increases with the gap

spacing and the electrode diameter [31]. In the same publication, it was reported that the number and size of bubbles as the weak points to cause breakdown can be reduced by high pressures in SLN<sub>2</sub>. Similarly the low temperatures contribute to reduce the number and size of bubbles. The question as to whether the size effect is due more to the electrode area or to the stressed volume in SLN<sub>2</sub>, was treated in [21, 32]. Experimental results revealed that the breakdown mechanism changed from an area dominant to volume effective region at larger electrode configurations in SLN<sub>2</sub> [32]. Such behavior was also noted in conventional insulating liquids [29, 30, 33, 34], i.e. that the influence of the stressed volume is more crucial with larger electrodes.

In the presence of bubbles, breakdown strength is reduced. In such a case, the breakdown voltage depends on the bubble volume. Relatively recent research indicated the dependence of the breakdown strength of SLN<sub>2</sub> on the bubble presence. On the other hand, in such a case, an increase of pressure may have a beneficial effect on the breakdown strength since higher pressures suppress vapor bridge formation avoiding thus a precipitous drop in breakdown strength [35]. Bubbles represent the weak link in SLN<sub>2</sub> since the discharge activity starts from them. In fact, Paschen's law is also valid at cryogenic temperatures making thus easier the calculation of inception voltages [36, 37]. Furthermore, as was reported in [38], the development of discharges in bubbles is a process of ionizations and hence an increase in pressure affects the quantity of micro-bubbles. Increasing pressure and reducing temperature may therefore offer an effective means to reduce the probability of inception of pre-breakdown streamer activity since the aforementioned quantities may influence streamer behavior and thus reduce the number of PD events from an anode tip [38]. Reducing temperature has as a result the condensation of existing bubbles, which in turn leads to a decrease of the electrically weak links [39]. A sudden increase of temperature may cause – in combination with the electric stress – the so-called bubble disturbance. The latter may reduce the voltage breakdown from 165kV<sub>peak</sub> to 80kV<sub>peak</sub> for a gap spacing of 8mm with a sphere-plane electrode arrangement [31, 40].

#### VII. INFLUENCE OF UNIFORM AND NON-UNIFORM ELECTRIC FIELDS

The applied electric field has a preponderant role in determining the breakdown strength of insulating materials and/or of the insulation in electrical apparatus [16]. Insulating materials at very low temperatures could not be an exception to the rule. Earlier work indicated that the breakdown voltage of liquid helium (He) and liquid nitrogen (LN<sub>2</sub>) is higher with uniform fields than with non-uniform fields [27]. The initial increase of breakdown voltage with the gap spacing for both the aforementioned liquids, even with a point-plane electrode arrangement, and the subsequent leveling-off may be due to the low latent heat of cryogenic liquids, which may lead to the creation of vapor around the point electrode [27]. Similar data to [27] were collected in [41], where authors observed that the impulse breakdown voltage is higher than the AC breakdown voltage for both sphere-plane and point-plane electrodes, with the former giving a much more distinct difference than the

latter. Agreeing qualitatively with [27], the leveling-off of the breakdown voltage with increasing the gap spacing – in a sphere-plane arrangement – was also reported in [42], where the leveling-off was interpreted with the stabilization of the maximum electric field at larger gaps. The importance of bubbles on breakdown strength with various electrode arrangements was pointed out in [42]. More generated bubbles lower significantly the AC breakdown strength with both sphere-sphere and with tape electrodes, their effect being more emphatic with the latter [42]. The applied electric field has a vital influence on streamer propagation. Streamer propagation is to a significant extent determined by the macroscopic electric field [17, 27]. Experimenting with sphere-plane electrodes and with pressures of 500kPa, it was shown that positive streamers have a higher velocity than negative streamers and thus, positive streamers result in lower breakdown strength [17, 27]. Such data were confirmed in yet another publication, where it was indicated that a transition from slow to fast positive streamer was observed at a threshold voltage below the breakdown voltage [43]. In [43], it was pointed out that at very low temperatures, with sphere-plane electrodes, and with impulse voltages there is a remarkable polarity effect [44], namely that at positive polarity a faster streamer was noticed whereas with negative polarity the phenomenon was slower. In this respect there is agreement between [45] and [17, 27].

#### VIII. ON THE DEGRADATION OF COMPOSITE INSULATING SYSTEMS

PD consist one of the major sources of degradation of insulating materials [46, 47]. In a non-uniform electrode system containing a disc of PTFE in LN<sub>2</sub>, an increase of the applied voltage led to an increase of the cumulative number of PD as well as of their maximum values [38]. An increase of pressure in such an insulating system from 100kPa to 400kPa, reduces the PD number, whereas a reduction of temperature does not have any remarkable effect on the slow negative streamers. On the contrary, a reduction in temperature causes a reduction of the fast positive streamers [38]. Further studies regarding the effect of pressure on the inception stress of a composite insulating system consisting of LN<sub>2</sub>/polypropylene, indicated a decrease of inception stress by about 13% - 40% when the pressure was reduced from 200kPa to 100kPa [48]. In a system consisting of LN<sub>2</sub>/polypropylene laminated paper (PPLP) with butt gaps, it was noted that the inception stress depended on the butt gap thickness and that thicker butt gaps resulted in a larger inception stress drop. This is probably due to the existing probability of weak points of the electrical insulation at butt gaps [49]. Regarding the role of butt gaps and their related parameters, there is a striking similarity between the observations of the above publication and those reported using conventional insulating systems [50, 51], i.e. that the butt gaps – if not well impregnated – may be the weak point of a composite insulation. PD current pulses recorded at pressures of 0.1MPa, 0.12MPa, 0.15MPa, and 0.2MPa, confirmed – in a more recent publication – the above observations, namely that with increasing pressure, the PD current pulses become smaller and smaller [21].

Solid insulators are used in combination with LN<sub>2</sub> for some HTS applications. Various solid insulating materials have been

tried, such as PPLP, Tyvek, Nomex, Kapton, PVA, PMMA, PVB and PA66. The breakdown strength of PPLP, Kapton and PA66 was shown to be superior to the breakdown strength of other materials [52]. In yet another study by the same group of authors, it was indicated that the breakdown strength of Kapton/LN<sub>2</sub>, polycarbonate/LN<sub>2</sub>, G10/LN<sub>2</sub> (G10 being fiberglass reinforced plastic) and polyetherimide/LN<sub>2</sub> depends on the frequency, and it indeed decreases with frequency [53]. Such a decrease is possibly due to the dissipation of space charge in the dielectric. According to earlier works, charge dissipation from charge traps depends on the frequency of the applied voltage leading to reduced partial discharge at low frequency [54, 55]. The properties of solid insulation at very low temperatures depend also on the additives as well as on the particular experimental conditions. For example, the HDPE presents a small increase of  $\tan\delta$  at 4.2K in comparison with the  $\tan\delta$  at 77K. Mechanical properties, such as the modulus of elasticity, are also most important since a combination of electrical and mechanical stresses may cause cracks [56-60].

#### IX. DISCUSSION AND PROPOSALS FOR FUTURE RESEARCH

Regarding HTS applications, it can be said that liquid nitrogen is an excellent medium for high voltage apparatus. It acts as both insulating and cooling medium for many HTS applications. It also has the advantage of low cost. It is chemically inert and thus the risk of fire is considerably reduced. One of the problems facing the HTS cables is that they must have high currents in order to have beneficial economic advantages for the companies and the end users. In the case of interruption of the superconducting state there may be a sudden failure, the latter may provoke a chain reaction which in turn may cause a generalized instability of the whole network. In such a case, the temperature of the cable may increase dramatically. That may have as a consequence that a considerable time interval will be required in order for the cable to reach its usual superconducting state. The remedy of a larger number of lapped tapes is not feasible since that would require increasing costs. Consequently, the possible solution will be in improving the materials of such a cable. Improved HTS cable designs based on YBCO as well as a reduction of the cost of cooling systems are proposed [61, 62]. During the last decade, the cost of superconducting materials has been reduced by about 10% per annum [62] and it is hoped that this trend will continue. Furthermore - although the cost of a superconducting apparatus is still high in comparison with the cost of a conventional apparatus - a factor that has to be taken into account is that much lesser space is required to accommodate superconducting equipment [63, 64].

With respect to materials research, further work has to be performed regarding the surface discharges in LN<sub>2</sub> /polymeric insulation uniform electrode arrangements [65]. Moreover, very low frequency (VLF) measurements regarding the insulation at cryogenic temperatures may offer an effective diagnostic technique to assess early insulation degradation [66-68]. Partial discharges due to defects must be further explored with insulating systems at very low temperatures both w.r.t their mechanisms and classification, as was done with conventional polymers at normal temperature with isolated as well as multiple defects [69, 70]. It is needless to say that with

the advent of polymer nanocomposites, a new prospect opens in the field of the applications at very low temperatures [71-74]. Research in this direction has already been performed and the prospects of employing polymer nanocomposites has been discussed, although one gets the impression that more effort was put into clarifying their mechanical properties rather than their electrical properties [75-78]. Also more effort has to be made regarding the expected life time of insulating materials (and insulating systems) at very low temperatures, especially in the view of earlier works on electrical stressing of HTS lapped tape insulated model cables [79].

#### X. CONCLUSIONS

In the present paper, a short review is offered regarding some aspects of insulating materials at very low temperatures. Some factors affecting the dielectric behavior of these materials are discussed. Although the very low temperature technology presents certain advantages compared to conventional technologies, more work has to be done w.r.t. the improvement of materials at such temperatures and the economic feasibility of such endeavors.

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