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General radiation problems for insulating materials in future fusion devices

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Abstract

It is envisaged that ITER will come into operation early in the next century. This next step machine will present problems due to the radiation damage effects in the numerous insulator components. The effects on the electrical conductivity, optical and mechanical properties are discussed in the light of recent advances and results. Material successes for the ECRH windows are presented, together with reasons for concern in the NBI system due to enhanced gas conductivity.

1. Introduction

It is envisaged that early in the next century ITER will come into operation, and it is hoped that this intermediate next-step "technology" machine will bridge the gap between the present day large "physics" machines and the pre-commercial DEMO reactor. Although ITER will undoubtedly help to solve many of the problems which still remain in the field of plasma physics, it will also present additional operational and experimental problems due to radiation damage effects as a result of the intense radiation field from the "burning" plasma. This ignited plasma will give rise to a high energy neutron and gamma flux, extending well beyond the first wall, from which one foresees a serious materials problem which has to be solved. In the initial physics phase of ITER it is the radiation flux which will be of concern, whereas in the later technology phase both flux and fluence will play important roles as the radiation damage builds up in the materials. For structural metallic materials the problem of radiation damage is expected to be severe, although tolerable, only near to the first wall, however the problem facing the numerous insulating components is far more serious due to the necessity to maintain not only the mechanical, but also the far more sensitive physical properties intact.

Insulating materials will be required in a number of key systems ranging from heating and current drive to diagnostics, which affect not only the operation, but also the safety and control of the machine. The radiation field will modify to some degree all of the important material properties. Unfortunately in general these changes do not improve the materials. Some of the changes will be flux dependent, while others will be modified by the total fluence. Clearly the former flux dependent processes will be of concern from the on-set of operation of future next-step devices. The fluence dependent effects on the other hand are the important parameters affecting the component or material lifetime. The properties of concern which need to be considered for the many applications include electrical resistance, dielectric loss, optical absorption and emission, as well as thermal and mechanical properties. Several papers have been published discussing both general, and more recently, specific aspects of radiation damage in insulating materials for fusion applications, those most relevant to the present paper are included [1–14].

In this paper recent work on the degradation of the electrical, mechanical, and optical properties, together with the important advances in materials for ECRH windows, will be presented. A specific new problem due to radiation induced conductivity in the insulating gas for the NBI system will also be discussed. The important

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aspect of simulating the operating environment for the component or material under examination, is briefly mentioned, in reference to present experimental procedures.

2. Degradation of insulator electrical resistance

Electrical resistance, more generally discussed in terms of conductivity, is an important basic parameter for numerous systems and components including the NBI heating system, ICRH windows and supports, magnetic coils, feed-throughs and stand-offs, MI cables and wire insulation. The main candidate material for these applications is Al_2O_3, and is also the one which has been most extensively studied, both in the polycrystalline alumina form and as single crystal sapphire. At the present time four types of electrical degradation are recognised and being investigated, these are; Radiation Induced Conductivity (RIC), Radiation Induced Electrical Degradation (RIED), Surface Degradation, and Radiation Induced Electro-Motive Force (RIEMF).

Of these types of degradation, RIC was the first to be addressed in a fusion context, as this enhancement of the electrical conductivity is flux dependent and hence a possible cause for concern from the onset of operation of any fusion device. RIC has been studied for many years, and a sound theoretical understanding exists [15–19]. The first experiments carried out within a fusion application context, i.e. oxide materials, high dose rates and temperatures, gave an insight into the effects of dose rate, temperature, and material impurity, and established the well known relationship between the total electrical conductivity measured during irradiation and the ionizing dose rate [19–22]; $\sigma_{\text{total}} = \sigma_0 + KR^\delta$ where σ_0 is the conductivity in the absence of radiation, R the dose rate, and K and δ constants. Although $\delta \approx 1$, the detailed studies found temperature, dose and dose rate dependence in this parameter, with extreme values in certain cases ranging between 0.5 and 1.5, and a temperature dependence in K. At the present time extensive data is now available for materials irradiated with X-rays, γ-rays, electrons, protons, positive ions, and fission and 14 MeV neutrons. Many of the additional results, although in some cases limited to one temperature, and/or one dose rate, add confirmation to the earlier extended studies, but more importantly show that RIC is a function of the ionization, independent of the irradiating particle or source. With very few exceptions, all the data taken together over a range of dose rates from <1 to about 10^4 Gy/s shows $\delta \approx 1$, and lie within a narrow band with the spread in conductivity values at any given dose rate being about two orders of magnitude [11], see also for example K. Noda et al. [23], where 14 MeV neutron results are given together with a small selection of other RIC data. For all the RIC data available, due to the different experimental conditions it is difficult to draw any conclusions as to the reason for this spread in values. Data obtained from electron irradiations of different materials under identical conditions of dose rate and temperature (700 Gy/s, 450°C) gives an indication that the RIC is inversely proportional to the sample impurity content, as may be seen in Fig. 1 [24]. However it is important to remember that the type of defect (impurity) is important, as observed at lower temperatures and lower dose rates, and that the observation of an inverse proportionality should not be generalised. In the specific case of high temperatures the impurity induced traps just below the conduction band will be unstable. The RIC will then be governed more by deep traps and recombination centres, and may possibly be less sensitive to the specific impurity type. This can only be verified by laborious systematic RIC measurements.

Nevertheless despite such problems, from all the data available at the present time one can safely say that RIC is sufficiently "well understood" to allow this type of electrical degradation to be accommodated by the design, and that materials exist which give rise to electrical conductivities $\leqslant 10^{-6}$ S/m for ionizing dose rates of up to 10^4 Gy/s. One only expects possible problems or influence near the first wall.

In contrast to RIC, RIED is a more serious problem, not only from the point of increasing the electrical conductivity through a degradation of σ_0, but also because this type of degradation is still not fully understood, nor even is there general agreement as to whether RIED exists as a real volume degradation. The first report within a fusion context of radiation induced electrical degradation or RIED effect was for electron irradiated sapphire (Al_2O_3) and MgO [25], following which numerous experiments have been carried out to assess its possible relevance to fusion insulator applications [26] and references therein. These have been con-

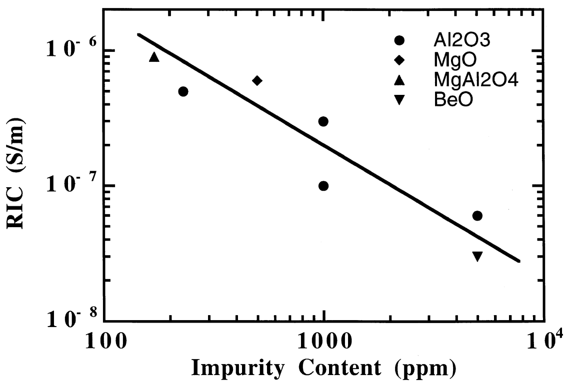

Fig. 1. RIC for different single and polycrystalline materials measured at 700 Gy/s, 450°C, plotted as a function of the estimated total impurity content. The line is of slope −1.

cerned with the effect of the applied electric field; DC or AC/RF and voltage threshold, the irradiation temperature, the dose rate, RIED experiments with protons, alphas, and fission neutrons, and other materials; $MgAl_2O_4$, AlN, and MgO insulated cable. Experiments have been carried out not only in vacuum, but more recently also in air and helium. Some experiments have not observed any RIED effect, or have observed enhanced surface conductivity. Extensive discussions on RIED have been held at two IEA Workshops [27,28], in an attempt to reconcile the conflicting results. The most recent of these workshops [28], reviewed the present state of RIED experiment results, and the reader is referred to this report for the numerous details. As far as electron irradiations are concerned, two research groups reported observation of bulk RIED, whereas two further groups did not. In contrast the reports for ion and neutron irradiations were all negative, with some previous reports of bulk RIED being reattributed to sample cracking (see later in Section 4 on mechanical degradation). Two tentative models based on aluminium colloid/gamma alumina formation, and on charge injection were discussed in an attempt to explain the electron RIED results, and are expected to be published soon. It was again pointed out that important factors such as material type differences and irradiation temperature, which could cause RIED not to be observed were not being taken into account [29]. However despite this, several recent experiments, also reported at the Workshop, have been performed at temperatures either near room temperature or above 600°C, considerably outside the expected temperature range. The most recently completed in-reactor RIED experiment in HFIR at ORNL [30] helps to throw light on the complex RIED problem, and in particular the possible material dependence. Initial results, as reported at the Workshop [28], indicated no significant increase in electrical conductivity for the 12 different samples. However further analysis has revealed moderate to substantial electrical degradation in some of the sapphire samples [31]. So clearly material type is an important parameter, and the high purity material (sapphire) appears to be more susceptible to degradation. One of the major difficulties for in-reactor experiments is the determination of σ_0, the conductivity in the absence of radiation, and its temperature behaviour. The use of nuclear heating and the residual reactor radiation level mean that changes in this parameter and its corresponding activation energy are not generally measured, although these are the main indicators, and RIED only becomes measurable when $\sigma_0 \geqslant KR^{\delta}$.

The specific problem of the radiation environment is worth mentioning. Most accelerator experiments to study electrical conductivity have been performed in vacuum, whereas for technical reasons in-reactor experiments have in general been carried out in helium. In this context it is interesting to note that one experiment in which sapphire was electron irradiated in air [32], showed a low saturation in the RIED degradation not observed in the vacuum experiments reported by Hodgson. In a similar way, the only two reactor experiments performed in vacuum [33,34] observed marked electrical degradation. Clearly the role of the irradiation environment in electrical degradation requires further study.

The importance of the radiation environment leads directly on to the problem of surface degradation. For the insulating components, surface degradation is just as serious as volume degradation, despite the purely academic distinction. Following reports of enhanced surface degradation in accelerator RIED experiments [35,36], it was suggested that the RIED degradation is not a real volume effect, but is caused by surface contamination. However two types of surface degradation have now been reported, that related to surface contamination caused by poor vacuum, sputtering, or evaporation [35,36], and real surface degradation of the material related to surface vacuum reduction and possibly impurity segregation [37,38]. Both forms can be serious, and are affected by the irradiation environment and ionizing radiation. However the real surface degradation effect is strongly material dependent, being observed in one type of alumina but not in another under identical irradiation conditions [37,38]. It is important to note that this type of degradation occurs in vacuum but not in air or helium.

Finally mention must be made of a further type of electrical degradation, RIEMF. Strictly speaking this is not a degradation, but an induced voltage which “degrades” the signal quality. It has been observed in MI cables during in-reactor experiments. The origin is still not at all clear and the question arises as to whether it is a real effect or due to experimental difficulties in an electrically noisy environment. However recent experiments indicate that the RIEMF is a real effect and can reach several volts or supply tens of microamps of current [30,39]. The voltage is possibly generated within the MI coaxial cable due to the difference in surface area per unit length between the central and outer conductor causing a charge build up near the central conductor, as observed in wire ionization chambers. Further work is still required to fully assess this phenomenon.

As concluded in the Cincinnati Workshop [28], while RIED itself may not be an issue for near term fusion devices such as ITER, the numerous experiments which have been carried out in relation to RIED have highlighted several additional technical problems such as surface conductivity, cracking, RIEMF, mineral insulated cable terminations, all of which require assessment to ensure that they can be accommodated in future fusion device design.

3. Advances in ECRH window materials

It is undoubtedly in the field of window materials for ECRH applications where the most notable progress has been made in recent years. An initial critical assessment of the problem showed that only materials with an adequate combination of low dielectric loss, high thermal conductivity, and mechanical strength could safely transmit the megawatts of RF power required [3,9]. It was soon demonstrated that sapphire, the ECRH window reference material, which has very low loss in the IC, LH, and EC frequency ranges, could only be employed at cryogenic temperatures and with a very low neutron tolerance level of about $\leqslant 10^{20}$ nm^{-2} [40]. Due to this marginal situation, work was started on possible alternatives to sapphire, which could provide windows for operation at frequencies above 150 GHz and at temperatures near to 20°C. Within this context, diamond and silicon are being studied, which in contrast to sapphire show decreasing loss with increasing frequency [41]. Grades of these two alternative materials are now available which show extremely low losses, in the range of 10^{-5} at room temperature, comparable with that of sapphire at 77 K.

The development of these two materials has been in completely opposite directions. On the one hand the initial high resistivity (HR) silicon was found to have very low loss but to be extremely radiation sensitive. Due to its perfection, electrons excited into the conduction band by ionizing radiation had very long lifetimes (no defect recombination sites) leading to high dielectric loss through the high electrical conductivity. On the other hand the initial CVD diamond material, almost black in colour, was found to have high loss due to the numerous defects in the material giving rise to polarization losses, but to be almost insensitive to ionizing radiation due to the extremely short lifetime of the electrons excited into the conduction band [42,43].

It was found that the high radiation sensitivity of silicon was remarkably reduced by electron irradiation through the introduction of recombination defects caused by the displacement damage, and also by Au doping, which has the same effect as pre-irradiation [42–45]. This reduction in the sensitivity may be seen in Fig. 2 [45]. Following these treatments no in-beam effects on the dielectric loss were observed at 40 GHz 0.7 Gy/s. The main limitation for silicon however comes from its small band gap of only 1.1 eV This gives rise to a substantial number of electrons being thermally excited into the conduction band at temperatures only slightly above room temperature leading to rapidly increasing dielectric loss [42].

Progress in CVD diamond for ECRH applications has been remarkable over the past few years, available samples going from black and irregular in shape, to almost transparent 1 mm thick 40 mm diameter discs, with

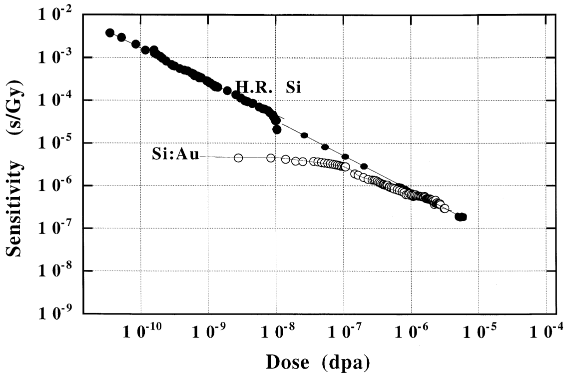

Fig. 2. The reduction of the radiation sensitivity of Si and Si:Au as a function of radiation dose.

the room temperature loss being comparable with sapphire at 77 K [46]. Measurements of the radiation effects on the loss in this improved material have found no in-beam effects at 18 GHz, 800 Gy/s during electron irradiation and no in-beam effects at 40 GHz, 0.7 Gy/s during X-irradiation [43,46]. As far as neutron irradiations are concerned, for diamond, Si, and Si:Au irradiated to 10^{20} nm^{-2} (about 10^{-5} dpa) at 50°C, little effect has been observed on the dielectric loss. In addition the thermal conductivity is unchanged [46]. Successful high power transmission tests have now been carried out on this highly promising material.

4. Degradation of mechanical properties

Early post-irradiation examination of the mechanical properties of aluminas indicated that significant degradation of the mechanical strength would only occur for radiation damage levels of the order of 1 dpa or above [47]. However recent work has found evidence for two types of enhanced degradation of the mechanical strength, enhanced implying degradation for $\ll 1$ dpa. The first of these is RIED associated. Sapphire and aluminas have been observed to become fragile, apparently due to internal stress following RIED degradation to damage levels of the order of 10^{-4} dpa [48,49]. This internal stress is most probably due to the formation of γ-alumina, which has been observed in TEM studies for RIED degraded sapphire [48]. It is not at all clear if γ-alumina formation is related to the reported cracking of Vitox alumina in a previously reported RIED degradation experiment [28,50]. However specific mechanical tests UBS on Deranox alumina electron irradiated with an electric field applied have so far failed to detect a change in the mechanical strength [51]. But it is important to point out that no significant electrical degradation was observed in the Deranox material in contrast to the nominally equal Vitox alumina which showed marked degradation. Such a difference in behaviour between these two grades was also noted by Möslang [28]. This problem clearly requires more attention.

The other type of enhanced mechanical degradation is related to sub-critical crack growth (SCCG), also in alumina [52]. A series of tests to determine the time-to-fracture of two types of aluminas (Deranox 975 and 995) held under different constant loads below the critical stress have shown quite clearly that the time-to-fracture is markedly changed when the ceramics are tested during irradiation at 1.5 Gy/s with ^{60}Co gammas. In the case of the 975 alumina a ten fold increase in the time-to-fracture was observed as may be seen in Fig. 3, while for the more pure 995 alumina a decrease by a factor of two was recorded. The authors note that in the case of the 975 alumina the gamma irradiation inhibits the crack growth and conclude that the effect is dependent on microstructural details of the alumina grain boundary phases. In fact these experiments show that we can have degradation (995) or improvement (975) in the mechanical properties depending on the alumina grade.

It is important to note that both types of degradation or modification in the mechanical strength are observed at very low radiation doses, $<10^{-4}$ dpa in the RIED case and $<10^6$ Gy for the SCCG, and are observed in situations of concurrent applied electric field or stress. Further work on the influence of concurrent stress and electric fields on mechanical strength is necessary to fully assess this potential problem.

5. Degradation of optical properties

Another area of recent concern is related to the effects of radiation on the optical properties of materials to be used as transmission components (windows, lenses,

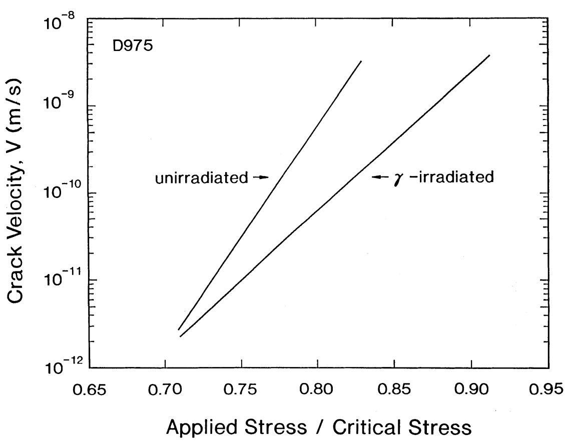

Fig. 3. Crack velocity decrease for gamma irradiated Deranox 975 alumina as a function of the relative applied stress.

and optical fibres) for the UV, visible, and IR wavelengths [53,54]. Radiation induced optical absorption and light emission (radioluminescence) impose severe limitations on the use of any optical material within a radiation field. SiO_2 and sapphire, present day ITER candidate materials for use in optical components for both diagnostic and remote handling systems, are not exempt from these limitations. For remote handling applications the optical components are expected to maintain their transmission properties under high levels of ionizing radiation (1–10 Gy/s) during many hundreds of h. For such applications radiation induced optical absorption imposes the main limitation. However in the case of diagnostic applications, in addition to a higher level of ionizing radiation (tens to hundreds Gy/s), the material will be subjected to atomic displacements of the order of 10^{-10} dpa/s. In this section two specific problems will be discussed, these are radioluminescence and background absorption in the visible and near infra-red, both of which require in situ measurements during irradiation. The general problems of the large absorption bands in the UV and visible region, introduced by displacement damage and related to oxygen vacancies, are not discussed as these are relatively easy to measure in post-irradiation studies. Considerable data now exists for this type of absorption in the main candidate materials SiO_2 and sapphire for high doses [55–58].

For the diagnostic applications radioluminescence, which like RIC is ionizing flux dependent, has recently been addressed and shown to be one of the main limitations for sapphire to fulfil the role of transmission component, making it extremely difficult to separate out the plasma emission from the window emission and absorption. While many studies have been carried out on luminescence phenomena in SiO_2 and sapphire, this problem has only recently been addressed in a quantitative way [59–62]. It was first shown for sapphire that the photon emission for a typical diagnostic window dose rate would be comparable with the photon emission from the plasma [59]. However work on the KU1 material provided by the Russian Federation for the ITER diagnostics radiation testing programme and classed as a quartz glass has shown that suitable materials do exist in which the radioluminescence can be reduced to a minimum [62]. Quantitative luminescence data comparing sapphire and two types of silica are given in Fig. 4, both of which show low radioluminescence. However one must keep in mind that with irradiation displacement dose the optical absorption in SiO_2 quickly renders this material opaque in the UV and visible range [55–58], and one may have to consider the use of sapphire over a very limited wavelength range near to 500 nm [62].

The LIDAR laser diagnostic system being considered for ITER will require very high quality transmission windows for the high power laser pulses at about 500

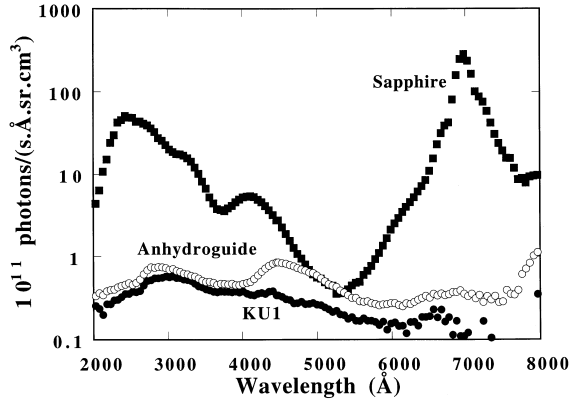

Fig. 4. Radioluminescence spectra for sapphire, Anhydroguide silica glass, and KU1 quartz glass at 20°C.

and 1000 nm. It is estimated that transmission losses of the order of 5% may cause problems with the window integrity due to laser damage. However the determination of such small decreases in the transmission corresponding to an optical density increase of only 0.02 are extremely difficult to measure by standard post-irradiation examination of irradiated material. Such measurements have to be performed in situ. Provisional results are now available which show that the absorption in the visible region for the KU1 material can give rise to up to 25% transmission loss for an 0.01 m thick window for total ionizing doses of less than 10 MGy, however the behaviour is strongly temperature dependent and complex, as may be seen in Fig. 5 [63].

6. RIC in the NBI gas

Finally a specific and unexpected radiation problem in the NBI system related to the insulating gas will be presented. In recent years considerable concern has been expressed about the possible radiation induced degradation of solid insulating materials under a fusion radiation environment, and by implication in those required for the ITER NBI accelerator system. However, in contrast little or no attention has been paid until very recently to the problem of the insulating gas which will be required around the NBI high voltage feed line, ion source and accelerator [64–68]. This gas, in the present design SF_6, will be in a radiation field of the order of 1 Gy/s due to the plasma and the NBI accelerator itself. The radiation will cause ionization in the gas, and hence an increase in the gas electrical conductivity. As this is a source of power loss due to the corresponding leakage current which in addition will produce heating and possibly breakdown, the radiation effect must be quantified and taken into account in the engineering design of the NBI system.

Present results for dry air, He, N_2/CO_2 mixture, N_2, CO_2, and SF_6 which have characterized the gas behaviour in terms of ionizing dose rate and electric field, show that the gas does not behave like a solid insulator, but that the leakage current (gas electrical conductivity) is a function of the gas volume due to the possibility of collecting all the generated charge carriers. For SF_6 at atmospheric pressure and 20°C a leakage current of about 0.05 A/Gy/s/m^3 was obtained. With such a high value the extrapolated implications for the 1 MV ITER NBI system indicate that up to megawatts of power could be lost due to this radiation induced leakage current (Fig. 6). This finding has led to discussions on the possible use of vacuum insulation for the NBI ion source and high voltage feed line. Further work is going on in this field to reduce the uncertainties in the large extrapolations.

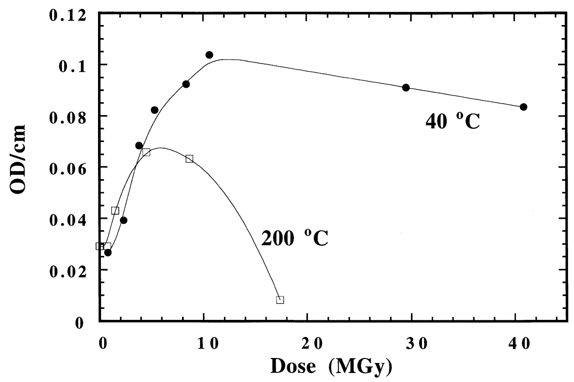

Fig. 5. Optical absorption changes at 500 nm in KU1 quartz glass as a function of dose for irradiation at 40°C and 200°C.

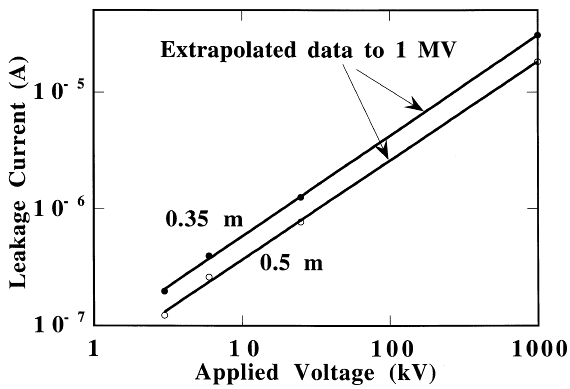

Fig. 6. Extrapolated leakage current in SF_6 at 1 Gy/s for two different electrode separations.

7. Conclusions

The important problems of electrical, mechanical, and optical degradation in insulating materials for next step fusion devices have been briefly presented, together with the most recent work on alternative materials for ECRH systems. Notable advances have been made in the classification of RIC for numerous materials, however the problem of RIED still remains to be fully understood. The importance of irradiation testing under relevant conditions of environment, mechanical strain and electric field have been highlighted with respect to RIED, surface effects, and mechanical property changes. The neglected aspect of gas insulation for NBI systems has been discussed and shown to be a serious problem which may lead to megawatts of power loss in the gas.

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