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Temperature and radiation hardness of polycarbonate capacitors

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The aim of this paper is to examine the influence of temperature raising, as well as neutron and gamma irradiation on the dissipation factor (loss tangent, tgδ) and capacitance (C) of capacitors with polycarbonate dielectric. Operation of capacitors subject to extreme conditions, such as high temperature and in the presence of ionizing radiation fields, is of special concern in military industry and space technology. The obtained results show that the rise of temperature has a considerable influence on the loss tangent of these capacitors, while exposure to a mixed neutron and gamma radiation field causes a decrease of their capacitance.

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1. Introduction

Dielectrics used as capacitor insulators have a wide range of physical and structural characteristics, which give rise to differences in their electrical behaviour. Some of the most important characteristics of a capacitor dielectric are its polarization properties, electrical conductivity, power losses, and dielectric strength (maximum electric field strength it can withstand without breaking down). Among the factors which may influence these characteristics are the intensity and frequency of applied electric field, humidity, duration of exploitation, mechanical deformation, temperature and exposure to radiation.

The influence of ionizing radiation on the reliability of dielectric materials used for insulating sheathing of communication and power cables is negligible compared to the influence of other factors, such as temperature, humidity, aging, and chemical reactivity. On the other hand, effects of ionizing radiation on dielectrics used as capacitor insulators may result in significant changes of capacitor performance within an electrical circuit [1,2].

2. Influence of temperature on capacitor characteristics

Temperature raising leads to a change in dimensions of capacitor plates and of the spacing between them. It also causes a change of the dielectric constant and loss tangent of the insulator. As the temperature of a polymer based dielectric material rises, its density decreases, which results in higher electron and ion mobility, as well as enhanced orientation of large polar segments of macromolecular chains [3]. All these effects may produce both reversible and permanent changes of capacitance in a capacitor with polycarbonate dielectric. Reversible changes are characterized by the capacitance temperature coefficient α_C, usually quoted in parts per million per degree Celsius (ppm/°C), which can be expressed as a sum of three coefficients, each reflecting one of the stated phenomena:

$$\alpha_C = \alpha_\varepsilon + \alpha_s - \alpha_d \tag{1}$$

where α_ε is the dielectric constant temperature coefficient, α_s effective electrode surface temperature coefficient, and α_d gap-size (plate distance) temperature coefficient. The gap-size contribution is included with a negative sign, since capacitance is inversely proportional to the distance between plates. Self-compensation of temperature coefficients referring to geometrical changes is commonly employed during capacitor design, by choosing plate materials and dimensions so that $\alpha_s = \alpha_d$. This reduces temperature dependence of capacitance to the thermally induced changes in dielectric, which are also responsible for changes of the loss tangent value [4].

3. Influence of ionizing radiation on polymer dielectrics

Fig.1 gives a schematic display of ionizing radiation effects in polymer materials [5]. The influence can be divided into two phases. In the first phase, gamma photons give rise to secondary electrons by way of Compton scattering and photoelectric absorption. Neutron induced nuclear reactions give rise to alpha particles, which go on to produce delta electrons along their tracks. Secondary electrons produced by gamma and neutron radiation further interact with polymer macromolecules, causing their ionization and excitation.

In the second phase, relaxation of excited molecules and locally formed ionization clusters results in formation of large amounts of free radicals. Highly reactive free radicals cause destruction of polymer chains, by either chain scission (random rupturing of bonds) or cross-linking (formation of large three-dimensional molecular networks). As a result of chain scission, low-molecular-weight fragments, gas evolution, and unsaturated bonds may appear [6,7].

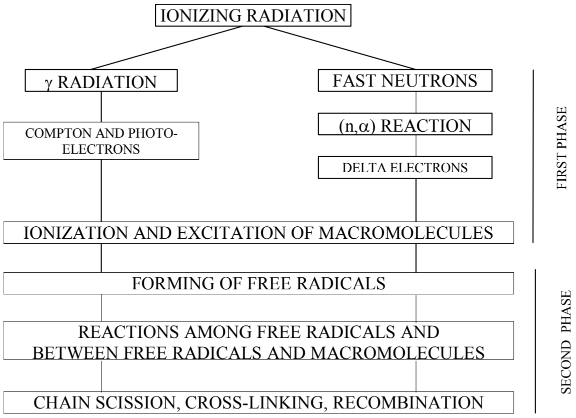

Fig. 1. Ionizing radiation effects in polymer materials.

Free radicals formed during irradiation can survive for several weeks before recombining. The ratio of resultant recombination, cross-linking, and chain scission depends on the chemical composition and morphology of the polymer, the total radiation dose absorbed, and the rate at which the dose was deposited. Unlike polytetrafluoroethylene, polypropylene, or polymethyl-methacrylates, the physical properties of polycarbonate remain virtually unchanged at low absorbed doses ($< 10^5$ Gy). This occurs despite the relatively large chemical transformations and extremely long lifetimes of the formed free radicals [8-10].

Stabilization techniques, aiming to increase radiation tolerance of polymers, make use of compounds acting as free radical and electron scavengers. Free radical scavengers are capable of generating a large number of hydrogen radicals, which terminate free radicals produced in the polymer by radiation. Electron scavengers degenerate into radical/ion pairs upon colliding with a high-energy electron. This mechanism reduces the energy of secondary electrons, and can thereby reduce the damage caused by radiation in polymers. The combination of both electron and radical scavengers in polymers produces a complete protective effect. Although electron scavengers help stabilize the polymer macromolecule, they nevertheless cannot prevent the formation of radicals. These radicals must still be terminated with the help of free radical scavengers [11].

Many important physical and chemical properties of polymers can be modified with radiation. Among these are molecular weight, chain length, entanglement, polydispersity, branching, and chain termination. These structural changes alter electrical properties of polymers used as dielectrics in capacitors, affecting the loss tangent and capacitance [12,13].

4. Experimental procedure and results

Polycarbonate dielectric capacitors used for both temperature and radiation investigations had the customary extended foil design, with metallized electrodes.

Temperature dependence investigations were performed on capacitors with 0.33 μF nominal capacitance. Capacitance and loss tangent values were determined in the temperature range from -25 °C to 100 °C. The object was to investigate the change of capacitance (C) and loss tangent (tgδ) caused by the temperature rise. At first, the dependence of tgδ on temperature was measured at 10 kHz frequency of the applied voltage. The obtained results are shown in Fig. 2. The graph shows two regions with elevated tgδ values, at low and high temperatures.

After keeping the capacitor at 100 °C for 30 minutes, the dependence of tgδ on the frequency of the applied voltage was measured, and then compared to the same dependence measured at 25 °C. Results are presented in Fig. 3 for the frequency range 100 Hz - 100 kHz. The graph points to a higher loss tangent in the capacitor heated to 100 °C at all frequencies.

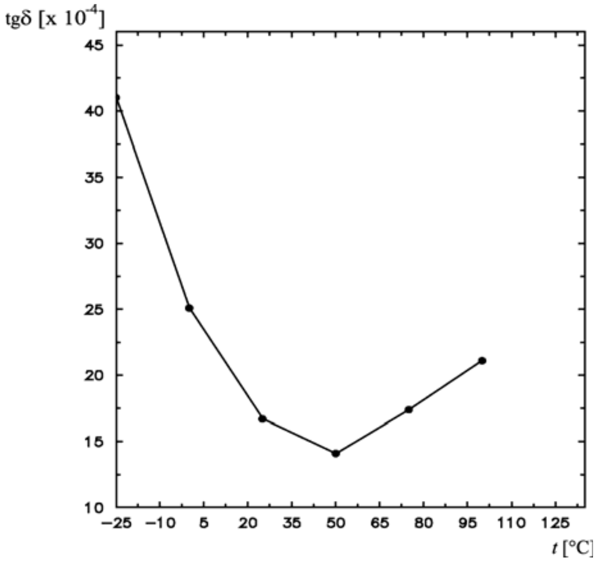

Fig. 2. Dependence of the loss tangent on temperature at 10 kHz frequency of the applied voltage.

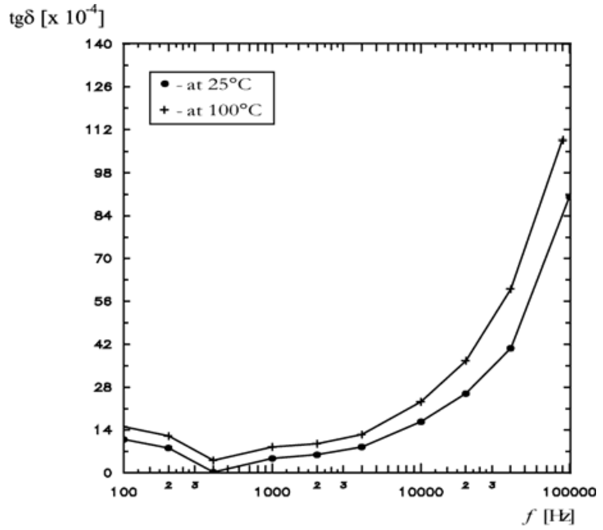

Fig. 3. Dependence of the loss tangent on frequency at 25°C and 100°C.

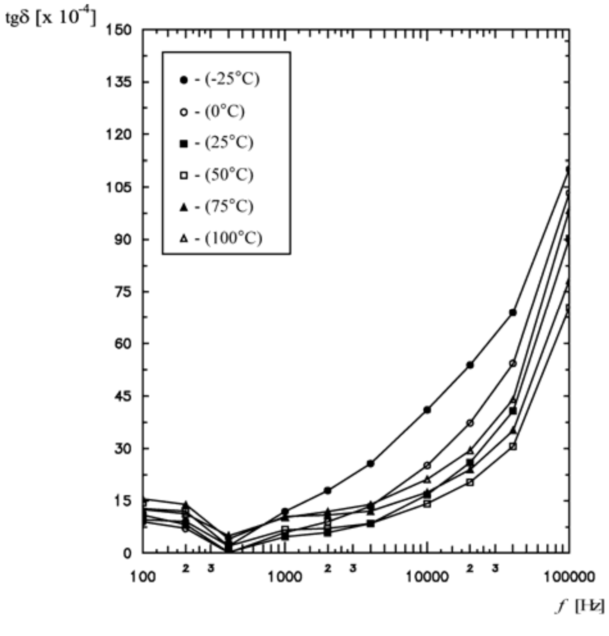

Fig.4. Dependence of the loss tangent on frequency at -25°C, 0°C, 25°C, 50°C, 75°C, and 100°C.

Finally, the graph in Fig. 4 shows tgδ versus frequency in the 100 Hz - 100 kHz range, for six temperature values (-25°C, 0°C, 25°C, 50°C, 75°C, and 100°C). High values of tgδ at low temperatures, previously observed at 10 kHz (Fig.2), are present only at frequencies higher than ~ 1 kHz. On the other hand, the rise of tgδ at high temperatures is detected throughout the whole frequency range.

Capacitance of the investigated polycarbonate capacitor exhibited a negligible rise with temperature. At -25°C the capacitance was 339.35 pF, while at 100°C its value rose only to 345.1 pF. The frequency of the applied voltage was 100 kHz.

Polycarbonate capacitors used for radiation tolerance investigation had the nominal capacitance of 1 μF, nominal impedance of 10^{14} Ω, and nominal loss tangent of 15×10^{-4}. The capacitors were exposed to a mixed neutron and gamma (n+γ) field from a ^{252}Cf source, encapsulated in the form of Cf_2O_3. The mass of the used ^{252}Cf radionuclide was 2.265 μg, its specific neutron and gamma emission rates $2.34 \cdot 10^6$ (μg s)$^{-1}$ and $5.3 \cdot 10^9$ (μg s)$^{-1}$, respectively. All post-irradiation measurements of the loss tangent and capacitance were performed at the fixed voltage frequency of 100 kHz. Average neutron energy of the ^{252}Cf source is 2.14 MeV, and average gamma photon energy is 0.88 MeV.

Table 1. Values of neutron (Φ_n) and gamma (Φ_γ) fluences used for polycarbonate capacitor irradiation

n_Φ	Φ_n [10^{10} cm^{-2}]	Φ_γ [10^{13} cm^{-2}]
1	2.79	6.8
2	5.59	13.6
3	8.37	20.4

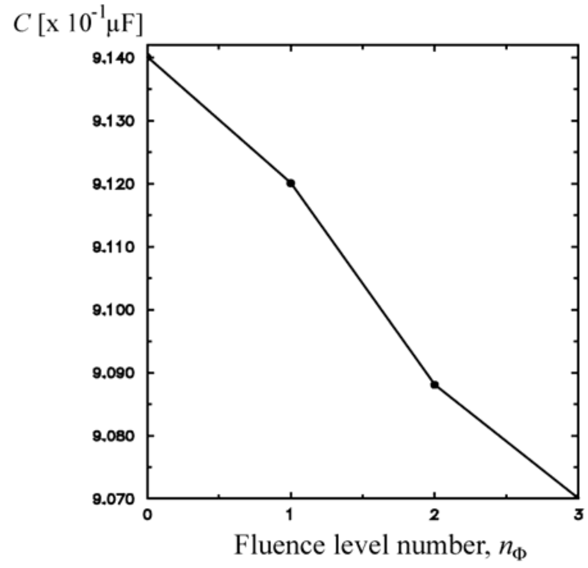

Fig. 5. The capacitance of the polycarbonate capacitor versus the fluence level number.

Polycarbonate capacitors were exposed to three different levels of total neutron and gamma fluences (Φ_n and Φ_γ respectively), given in Table 1 in ascending order. Each fluence level is marked by a number (n_Φ) shown in the first column. Values of capacitance measured immediately after irradiation are presented in Fig.5. The graph shows the decrease of the measured capacitance with fluence level number n_Φ, i.e. with the rise of fluence. The capacitance was measured again 120 hours after irradiation. The obtained values were all close to the nominal capacitance, which leads to the conclusion that the effects of radiation had meanwhile been reversed.

As opposed to capacitance, no measurable influence of $n_+\gamma$ radiation on the loss tangent has been detected.

6. Discussion

The observed high values of the loss tangent at low temperatures can be explained by the polarization of small polar segments of the polycarbonate macromolecule. The rise of the loss tangent at high temperatures, within the temperature range used, is caused by the polarization of large segments of the macromolecule. The performed temperature investigations lead to irreversible changes of capacitors' characteristics [3].

The decrease of capacitance of polycarbonate capacitors when exposed to $n_+\gamma$ radiation can be explained by the formation of ionic structures inside the dielectric volume. A higher concentration of ions in the dielectric causes a greater influence of ionic polarization on the dielectric constant of the material, leading to the decrease of capacitance [3]. In addition, local electric field of ionic structures performs partial screening of the capacitor's electric field, albeit the change in the capacitance attributed to the screening is relatively small. Permanent presence of ionic structures inside the dielectric can cause its dielectric strength to decrease and thus accelerate the aging of the capacitor [14].

Reversibility of the observed radiation effects is a result of recombination processes inside the polycarbonate dielectric. The fact that the effects were reversed after a period of time shows that the absorbed doses were not high enough to produce permanent changes in the molecular structure of the dielectric.

7. Conclusions

Results of examining temperature and radiation hardness of capacitors with solid polycarbonate dielectrics were presented in this paper. It was shown that the rise of temperature has a considerable influence on the loss tangent of these capacitors, while exposure to a mixed neutron and gamma radiation field causes a decrease of their capacitance. Radiation effects in the polycarbonate insulator were reversible at fluence levels used for this paper, but may lead to permanent degradation at higher doses, causing accelerated aging of capacitors. Further investigation should address the combined effects of high temperature and radiation exposure, as well as stabilization techniques for radiation hardening of polycarbonate dielectrics.

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