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Aging of organic materials around high-energy particle accelerators

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Abstract

Around particle accelerators used for fundamental research on the basic structure of matter, materials and components are exposed to ionizing radiation caused by beam losses in the proton machines and by synchrotron radiation in the lepton machines. Furthermore, with the high-energy and high-intensity collisions produced from future colliders, radiation damage is also to be expected in particle-physics detectors. Therefore, for a safe and reliable operation, the radiation aging of most of the components has to be assessed prior to their selection. An extensive radiation-damage test program has been carried out at CERN for decades on a routine basis and many results have been published. The tests have mainly concentrated on magnet-coil insulations and cable-insulating materials; they are carried out in accordance with the IEC 544 standard which defines the mechanical tests to be performed and the methods of degradation evaluation. The mechanical tests are also used to assess the degradation of composite structural materials. Moreover, electrical properties of high-voltage insulations and optical properties of organic scintillators and wave guides have also been studied. Our long-term experience has pointed out many parameters to be taken into account for the estimate of the lifetime of components in the radiation environment of our accelerators. One of the main parameters is the dose-rate effect, but the influence of other parameters has sometimes to be taken into account.

Keywords: Radiation; Dose-rate; Aging; Polymer; Organic

1. Introduction

For fundamental research on the basic structure of matter, the European Organization for Nuclear Research (CERN) operates several high-energy particle accelerators and storage rings. The first one, the 28 GeV Proton Synchrotron (PS), was brought into operation in 1959 and is still accelerating many types of particles today. The Super Proton Synchrotron (SPS) was built in the seventies and accelerates mainly protons up to the energy of 450 GeV. These accelerators eject their primary beams towards fixed targets where exotic particles are produced. The Low-Energy Antiproton Ring (LEAR) provides calibrated antiproton beams to physicists. The most recent machine, the Large Electron–Positron Collider (LEP) came into operation in 1989, it has a circumference of 27 km. The next one will be the Large Hadron Collider (LHC) located in the same tunnel as the LEP machine. Its superconducting magnets will operate at liquid-helium temperature and will enable the acceleration of protons up to the energy of 7 TeV; it will be the largest and the most powerful

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particle accelerator in the world. The LEP and the LHC are colliders which (will) provoke head-on collisions between primary particle beams circulating in opposite directions. From the collisions between stable particles, exotic unstable particles are produced. The study of their characteristics and behavior allows to understand the fundamental laws of physics.

The most important components of these machines are the electro-magnets to keep the beams on their circular orbits and to focus them. The magnet coils are usually insulated with epoxy-based laminates. These magnets as well as all other components have to be powered and controlled via hundreds of kilometers of electric cables.

All materials and components are exposed to ionizing radiation caused by beam losses in the proton accelerators and by synchrotron radiation in LEP. Furthermore, with the high-energy, high-intensity collisions produced from LHC, radiation damage is also to be expected in particle-physics detectors. Therefore, for a safe and reliable operation, the radiation behavior of most of the components has to be tested prior to their selection.

From the very beginning of CERN, attention has been given to the effects of ionizing radiation on components, notably on polymeric materials; Radiation-resistance tests have been performed on a routine basis on magnet-coil insulations and cable-insulating materials. The results are presented for a large variety of commercially available compounds and have been published in the form of catalogues [1].

2. Aging of polymeric materials

2.1. Definition

Usually, the properties of a material or of a component are not stable with time. Aging is the irreversible loss of properties of a material submitted to its environment. This permanent loss of property, which has to be distinguished from the transient change due to a temporary external stress, always results from a change in the intimate physico-chemical structure.

2.2. Mechanisms and factors of aging

There are several factors of aging, all of them depending on the environment. Some may seem to be inherent to the materials which change (evolve) with time because they are not in a stable thermodynamic state. When materials are used as components, they are subject to environmental conditions, and they inevitably age.

The mechanisms of aging depend on the material type. In comparison with metals and ceramics, which are mainly sensitive to corrosion, polymers are more sensitive to aging: they are more affected by external chemical factors such as oxidants (water, oxygen, ozone...) and pollutants including bacteria, and physical factors such as temperature (and temperature changes), mechanical stresses (including abrasion and fatigue), electrical and/or magnetic fields, light (mainly UV), microwaves, ultrasonic, ionizing radiation, etc. [2–4]. In any material, internal defects such as impurities, grains and cavities will usually facilitate the action of the other factors. During the transformation process, the temperature is raised above a phase transition, and hydroperoxydes which will be potential initiators for further oxidative degradation are produced [5]. The proper choice of polymerization and transformation processes is crucial for the aging.

As said above, aging results from physico-chemical modifications of the material. In polymers, these modifications are mainly due to chain scissions, crosslinkings and changes of side groups. Chain scission also called degradation is the random cutting of the main chains, which results in a decrease of the mean molecular weight. Degradation may also result from depolymerization or "unzipping" of the main chain which leads to products similar to the parent monomer. Crosslinking creates new links which may form a three dimensional network in thermoplastics. Changes of side groups include cyclisation and elimination of low molecular units leading to the formation of unsaturations and gas evolution. Polymers are also subject to phase changes; the crystalline phase of plastics may be destroyed by irradiation; on the other hand, the energy brought by radiation may anneal amorphous plastics in which crystallites will be produced. In polymeric materials, additives such as organic or mineral fillers, and stabilizers such as

antioxidant and UV protectors, may also be modified and/or eliminated with time; they usually change the mechanisms and the rate of aging.

2.3. Critical properties, problem of monitoring

If aging leads to physico-chemical changes in the polymers, its evolution can be monitored by various techniques such as differential scanning calorimetry, infra-red spectroscopy, electron paramagnetic spectroscopy, nuclear magnetic spectroscopy, molecular weight measurements, gel fraction measurements, etc. Usually, the manufacturers will study the aging of their materials by some of these techniques, in order to understand the mechanisms, to avoid them and to produce better compounds.

From the user's point of view, having identified the aging phenomena is not enough; to have an impact on reliability and safe use, it must also be possible to follow-up aging by the measurement of one or several macroscopic properties. As materials are used as (part of) components, they have a (or more) specific function(s) requiring specific properties. It seems therefore logical to test the mechanical properties of a structural material, the electrical properties of an insulator, the optical properties of an optical components, and so forth. But this is not always neither the easiest nor the best way. Experience has shown that it is often possible to assess the aging of an insulator from the degradation of its mechanical properties. In fact, the standard 544 of the International Electrotechnical Commission recommends to measure the degradation of insulators from mechanical tests [6].

The IAEA has set up a program to assess the aging of cables in nuclear power plants. It also recommends mechanical tests to be performed, but notes that having studied the materials' behavior and their mechanisms of aging is only one part of a proper aging management program: a major need is the specific knowledge of the environmental conditions [7].

In the accelerator tunnels at CERN, the temperature is usually quite constant and close to normal conditions i.e. 18°C to 25°C. As the tunnels are ventilated with outside air, the relative humidity may vary from 30% to 90%. As radiation levels may be as high as several tens of grays per hour, leading to doses of megagrays (MGy) per year, they are the main factor of aging. A complete program dose measurements has been set-up for many years [8].

3. Results of radiation-aging tests

3.1. Selection of insulating materials at CERN

As said in the introduction, numerous radiation-resistance tests are carried out at CERN on a routine basis, in accordance with the recommendations of the IEC 544 standard. Flexural tests are carried out on rigid insulators (thermosets, varnish) and tensile tests on flexible insulators (mainly thermoplastics and rubbers used for cable insulation). The standard also recommends that accelerated tests be performed after irradiations at dose rates greater than 1 Gy/s, and that more representative tests be performed after aging conditions as close as possible to in-life conditions.

3.1.1. Magnet-coil insulations

The magnet coils are located very close to the metallic vacuum chamber where the high-energy beams circulate, and therefore close to the source of radiation. For the insulation of these coils, aromatic epoxy-resins (type bisphenol A) with specially formulated anhydride hardener and accelerator are used [9]. Over the years, several manufacturers have gained experience, the formulations have changed somewhat, and it is nowadays possible to find radiation-resistant epoxy-resins (up to tens of megagrays). Well-prepared laminates and composites may resist

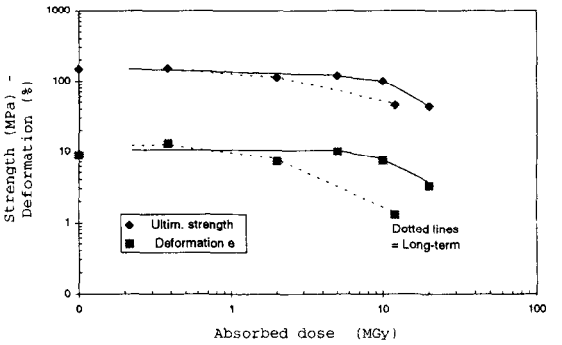


Fig. 1. Radiation degradation in epoxy R 423.

doses a factor 10 higher [1,9,10]. It has been shown that the IEC recommendations are valid: the degradation is usually more pronounced for the mechanical properties than for the electrical properties [11].

The "*dose-rate effect*" on an epoxy compound (R 423) is shown on Fig. 1; for the same total absorbed dose, the degradation of the mechanical properties is more pronounced after a long-term irradiation (0.5 kGy/h) than after an accelerated one (200 kGy/h). This is often observed in heavily filled resins [12], but most of the time, no dose-rate effect can be observed in the best glass-fiber-reinforced composites since the degradation has not started at the maximum dose obtained during the test [10,13].

3.1.2. Cable insulating materials

Since the eighties, for the design of the LEP machine, it was decided that for safety reasons in case of fire, the materials selected for the construction had to be fire-retardant and halogen-free (in case of a fire, the resulting smoke must be light, of low toxicity and noncorrosive). As a result of this decision, some commonly used materials (e.g. PVC, chlorosulfinated polyethylene (CSP), and other halogenated and sulfur containing compounds) had to be excluded. The replacement of the halogenated materials was achieved in two steps: EPR/EPDM-based rubber compounds were used for medium- and high-voltage power cables, and polyolefin-based compounds were used for instrumentation and control cables.

Mechanical tests carried out after either accelerated irradiations or after long-term irradiations have shown that polyolefin-based compounds usually present an important *dose-rate effect* [14–17]. It is recognized that the dose-rate effect is related to the presence of oxygen (and/or other oxidants). During a long irradiation, oxygen diffuses into the polymer which then degrades via radio-oxidative processes. Fig. 2 shows the strong dose-rate effect (0.1 kGy/h compared to 4 kGy/h) observed in the degradation of the mechanical properties (mainly the elongation at break) of a polyolefin-based cable jacketing material. This effect is often combined with a *temperature effect* [18–20], and the techniques cited in [5] may be used to monitor the oxidative degradation.

A few years ago, an extensive program was carried out at CERN to estimate the influence of the

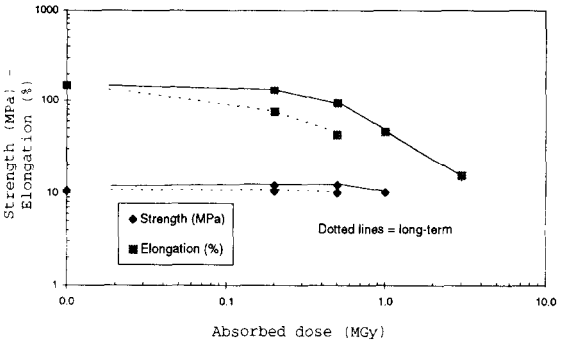


Fig. 2. Radiation degradation in polyolefin C 1037.

type of radiation on the cable degradation. It appears from the results that, except from the dose-rate effect, the degradation of cable insulations does not depend on the radiation type but only on the absorbed dose [21].

3.2. Other polymeric materials, other aging factors

Around particle accelerators and in complex particle-physics detectors, polymer-based materials are also used as pipes, hoses, spacers, covers, seals, foams, paints, small fixing parts such as cable ties, adhesive tapes and glues [1,21,22]. Most of these materials present an increased sensitivity to life irradiation compared to accelerated irradiations. For example, at doses between 0.5 and 1 MGy, the mechanical properties of rigid thermoplastics such as XL-polystyrene, polycarbonates and PMMA are about a factor of 2–4 lower after an irradiation at 100 Gy/h than after an accelerated irradiation at kGy/h [23].

Like many other organic materials, elastomeric seals also present a *temperature effect*; their degradation is usually more pronounced at higher service temperature. Models to predict their lifetime in service environments have been elaborated [24].

In view of the operation at liquid-helium temperature of the future LHC, a study has been made to assess the behavior of insulating and structural materials after irradiation at *cryogenic temperature*. The comparison of the results with the ones obtained at room temperature (RT) have shown that there is no significant influence of the irradiation temperature on the radiation degradation of thermosets and com-

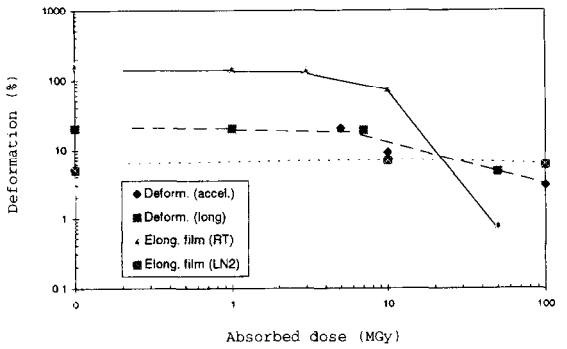


Fig. 3. Radiation behavior of polyether-ether-ketone.

posites, while the degradation of plastic films is even less severe as they are protected against oxidation [25].

The effect of oxidation on the radiation behavior of PEEK is shown on Fig. 3 from two different experiments. On one side, the reduction of the ultimate deformation of a bulk material (4 mm thick), where no dose-rate effect is observed, is less pronounced than the reduction of the ultimate elongation of a thin film (125 μm). On the other hand, no oxidative degradation of the film occurs in liquid nitrogen (LN2).

Structural materials are usually qualified from the values of their young's modulus, their strength, and/or their limit of elasticity. The flexural test is a representative one. Compression strength, impact strength and hardness may also be of concern. The physical dimensions (shrinkage or swelling) of the sample or components may also be checked. For these materials, the synergy between irradiation and *mechanical stress* has to be investigated. Some experiments have shown that rubbers and composites under stress are more sensitive to radiation. Also, carbon-fiber reinforced composites irradiated under stress degrade at lower radiation dose than unstressed samples. This effect is more marked for RT irradiation than for cryogenic irradiations. The enhanced degradation is more obvious in a composite with a toughened matrix (epoxy + plastic) [26].

The optical properties of mineral or organic glasses are in general more sensitive to aging than their mechanical properties and the former are also easier to measure. Very strong synergistic effects between radiation and other parameters are observed in plastic scintillators used for particle detection [27,28], translucent plastics used as wavelength shifters and light guides [29,30], as well as in optical fibers [31]. In the LEP, one optical cable is installed in the central water-drain, under 40 cm of concrete. The annual absorbed dose is therefore kept below one gray, and no radiation damage is observed up to now.

4. Conclusion

At CERN, for over 30 years, emphasis has been put on the proper selection of insulators to be used in the radiation environment around particle accelerators. A particular attention has been brought on the dose-rate effect study.

The good knowledge of the absorbed doses and the environmental conditions in the accelerator tunnels and high-radiation areas, combined with the knowledge of the materials' behavior, allows us to assess the lifetime of the selected components in our accelerator.

In the future large particle detectors, where more organic composites will be used as structural materials, other synergetic effects (mainly mechanical stresses, humidity and temperature gradients) have to be taken into account, and will be studied further.

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