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Material Properties

Comparative study on degradation of ethylene-propylene rubber for nuclear cables from gamma and beta irradiation

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ABSTRACT

Considering safety is the priority concern of nuclear power plants, equipment qualification testing of the nuclear components manufactured should be paid special attention to. Thereinto, equivalent conversion (1:1) from the absorbed beta doses to gamma doses is a rule of thumb for irradiation qualification testing of the polymers used as nuclear cables, however whether it is reasonable and applicable to Chinese domestic polymers still requires investigation. In this paper, both gamma and beta irradiation testing with the actual dose rates and total absorbed doses in China Advanced Passive (CAP) series nuclear power plant was performed upon one domestically manufactured ethylene-propylene rubber intended for nuclear cable insulation in China. The mechanical and the electrical properties before and after irradiation were measured to compare the extent and the trend of degradation between the two irradiation types, and related oxidation degradation mechanism especially its attenuation along the thickness was quantitatively addressed.

1. Introduction

Safety, the priority concern of nuclear power plants, is the capability of structures, systems and components (SSCs) to perform safety-related functions under both normal and accidental conditions against radiation risks [1], and is commonly credited by equipment qualification (EQ) and maintained by ageing management (AM) [2], thereby premature failure incidents could be prevented and conceived economic benefits could be ensured. Fundamentally, the precondition of safety relies on the reliability of materials. In practice, besides failure analysis of nuclear components with finished materials during operation [3–6], accelerated ageing testing [7–10] of raw materials under simulated service conditions before manufacturing is the other effective approach for improvement of materials reliability and longevity, among which irradiation is the characteristic factor to be taken into account.

As a matter of fact, ionizing irradiation degradation of polymers used in nuclear power plants (predominantly applied to the nuclear cables) has been attracting expansive researches since several decades ago [11–14], and effects including irradiation types [15,16], absorbed doses [17,18], dose rates [19], temperatures [20–22], and their synergisms [23–26] have been investigated. Thereinto, beta (β) irradiation testing is used to represent the post-accident environment in nuclear power plants, and is usually realized by electron beam accelerator. However, the dose rates of traditional electron beam accelerators are more than 10,000 kGy/h [27–29], at least three orders of magnitude higher than the actual β irradiation dose rates after design basis events (DBE) in containment [30]. Thus, the results of erstwhile β irradiation testing seem not be conservative enough if considering the dose rate effect in air, i.e. a lower dose rate with longer exposure time exerts severer degradation effect on polymers than a higher dose rate with shorter exposure time does when the total absorbed doses are same [31,32]. In this situation, the reasonability of equivalent conversion (1:1) from beta doses to gamma doses as a rule of thumb in EQ of nuclear components [33,34] deserves to be revalidated [35].

To this end, in addition to gamma (γ) irradiation by common ^{60}Co radioactive source, beta irradiation by a modified electron beam accelerator capable of generating dose rates from 10 kGy/h to 70 kGy/h (with patents) was conducted on one domestically

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manufactured ethylene-propylene rubber (EPR) product used for nuclear cable insulation of China Advanced Passive (CAP) series nuclear power plants in China. After irradiation testing, the mechanical and the electrical properties were measured to compare the different degradation effects from gamma and beta irradiation, and relevant degradation mechanisms were investigated by means of scanning electron microscope (SEM), attenuated total reflection Fourier transform infrared spectroscopy (ATR FT-IR) and FT-IR microscope. The results indicated that the degradation effects from both gamma and beta irradiation with dose rates and total absorbed doses in same order of magnitude (~10 kGy/h, ~10^3 kGy) were close on this EPR product, verifying the classic 'equal dose – equal damage' approximation in theory and the equivalent conversion from beta doses to gamma dose in practice for EQ testing of nuclear components.

2. Experimental

2.1. Material

The ethylene-propylene rubber is the domestically manufactured commercial product intended for application as nuclear cable insulation of CAP series nuclear power plants, and was provided as 2 mm-thick sheet from Shanghai Nuclear Engineering Research & Design Institute (SNERDI) for investigation. Samples of type 2 dumb-bell in ISO 37-2011 standard [36] and round plate with diameter of 80 mm were cut from the EPR sheet for irradiation, and subsequently subjected to the mechanical and the electrical properties measurement respectively.

2.2. Irradiation testing

The ^{60}Co radioactive source and the patented electron beam accelerator (0.7–1.5 MeV) was utilized to perform gamma and beta irradiation testing respectively on the EPR samples in air in Shanghai Institute of Process Automation Instrumentation (SIPAI). The irradiation conditions, as listed in Table 1, were specified according to the gamma and the beta irradiation dose rates and total doses after DBE in containment of Advanced Passive 1000 (AP1000) nuclear power plants [37]. Samples exposed to gamma irradiation were vertically hung around the ^{60}Co source, while those exposed to beta irradiation were horizontally and directly placed on a stainless steel plate with a φ 420 mm effective irradiation area under the moving electron beam, and flipped over at the middle of the total exposure time for better homogeneousness.

2.3. Measurement

Before and after irradiation, the dumb-bell samples were tested for tensile strength at break (TS_b) and elongation at break (E_b) in accordance with the ISO 37-2011 standard via WDS-W-5kN electronic universal testing machine (Chengde Precision Testing Machine Co., Ltd.), and the number of test pieces was 7 under each irradiation condition; the round plate samples were measured for volume resistivity according to the IEC 62631-3-1 standard [38] by ZC36 megger (Shanghai No.6 Electric Meter Works Co., Ltd.), and the number of test pieces was 3 under each irradiation condition.

Table 1
Conditions of the gamma and the beta irradiation.

Irradiation type	Dose rate (kGy/h)	Exposure time (h)	Absorbed dose (kGy)
gamma (γ)	10	10	100
		50	500
		100	1000
		500	5000
		1000	10000
beta (β)	20	25	500
		50	1000
		100	2000
		200	4000
		250	5000

2.4. Characterization

In order to identify the degradation mechanisms, Nicolet Nexus 470 ATR FT-IR (Thermo Fisher Scientific) was utilized to detect the functional groups changes of the samples before and after irradiation. Also, the cross sections of the irradiated dumb-bell samples were observed under S-520 SEM (Hitachi), and were even scanned by Nicolet iN10 FT-IR microscope (Thermo Fisher Scientific) after being cut into 15-μm-thick slices via Shandon Finesse 325 manual microtome (Thermo Fisher Scientific) to investigate the oxidation extent gradient along the depth.

3. Results and discussion

3.1. Mechanical properties

It is widely accepted that elongation at break is the most appropriate and sensitive index for evaluating irradiation degradation of elastomers [39]. As is displayed in Fig. 1(a), E_b of EPR from both gamma and beta irradiation decrease with absorbed doses in a roughly exponential decay pattern, indicating similar degradation extents on EPR from these two irradiation types. In more detail, the 50% relative drops of the initial E_b values (commonly regarded as the end-point for cable insulation elastomers) locate in around 500 kGy, and the ultimate E_b values are less than 10% of the initial.

However, with respect to the tensile strength at break, although in general they increase with absorbed doses, one exception is at the 1000 kGy absorbed gamma doses, seen in Fig. 1(b), which decreases nearly 20% compared with the value at previous dose (500 kGy), and consequently the situation that the TS_b increase rate from gamma irradiation is higher than that from beta irradiation before this dose (0–500 kGy) inverts after it (1000–5000 kGy). Besides, it can be learned that the statistical dispersion of TS_b is not as good as that of E_b, verifying the appropriateness of the latter for degradation evaluation of elastomers even though they are obtained simultaneously by one tensile test.

3.2. Electrical properties

As shown in Fig. 2, volume resistivity from both irradiation types increase with absorbed doses in low dose ranges until reaching peaks (gamma at 100 kGy, beta at 500 kGy), and then decrease in a sharp manner (gamma irradiation) and a gradual manner (beta irradiation) respectively. In detail, after the peaks, the volume resistivity from gamma irradiation slightly change in the range of ~10^{12} Ω·m (one order of magnitude lower than the initial values) with absorbed doses, while those from beta irradiation are all in the same order of magnitude of the initial values. Considering the 10% end-point criteria (90% drop) for volume resistivity in the standard [31], the critical absorbed dose of gamma irradiation for EPR can thereby be determined as 500 kGy, conforming with the E_b results mentioned above; on the contrary, EPR begins 'failed' after 500 kGy absorbed dose of beta irradiation based on the E_b results, while it seems still qualified even after enduring 5000 kGy beta irradiation from the electrical point of view, coinciding with the conclusion in literature [40,41] that cable materials could retain normal electrical functions even if only ~5% of the initial E_b values was left.