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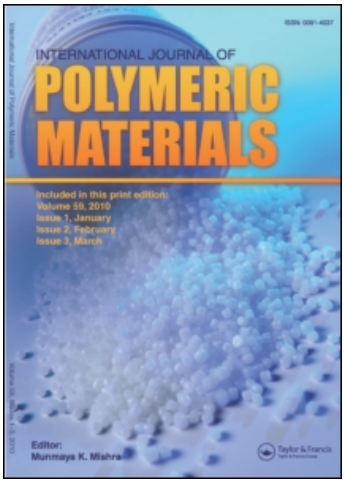

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Estimation of Heat- and Radiation Stability in Elastomers

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A new method to estimate the thermal and radiation stability of elastomers is presented.

KEY WORDS Thermal stability, radiation stability, elastomers

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Elastomers have to be of high stability (durability, τ) because of their long-term operation as parts of articles under aggressive conditions, the most important of which are high temperature and radiation. The GOST (Russian State Standard) methods of determining τ, widely practiced and based on the heat ageing of elastomers, are time- and material-consuming. They are unfit for high temperatures and make it impossible to estimate the contribution of different outside factors contributing to decreased rubber durability. A method developed by the present authors[1] lacks those drawbacks. It permits determination of the durability of elastomers 25–30 times faster, consumes a small quantity of material (a 120 × 140 × 1 mm rubber plate), makes it possible to extend the temperature range of τ determination and estimate the contribution of ionizing radiation to the decrease in stability. The new method allows thermo-oxidative destruction to be performed at temperatures similar to those used in the GOST methods but requires an additional superposition of the field of tensile stress which accelerates the destructive processes in samples and considerably decreases the time required for an estimation of the activation energy, U_0, and, consequently, determination of material durability. The method involves the common dependence (see Figure 1) which binds the rubber's durability and the activation energy of its thermo-oxidative destruction, E, which is found from the extensive experimental material on rubber durability by means of continuous thermal ageing. It also includes a procedure for U_0 estimation based on the temperature dependence of the ultimate tensile stress when thermo-oxidative destruction is accelerated by the superimposed field of mechanical forces.

In replicate tests using the method[1] and GOST 9713-86, the activation energies of the processes and the durability of 20 rubbers have been determined. The U_0 and E turned out to coincide in all cases to an accuracy of ± 2 kJ/mole while the

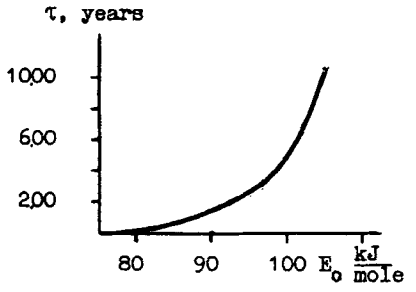

FIGURE 1 Experimental dependence of durability in air at 25°C on the activation energy of thermal ageing for rubbers of different polymeric bases and composition.

durability values correlate with one another. Therefore, the durability of elastomers can be determined according to the empirical formula:

$$\tau = 10^{(\alpha U_0 + \beta)} \cdot \exp(U_0/RT),$$

where $\alpha = -0.111 \pm 0.001$; $\beta = -3.687 \pm 0.05$.

The essence of the procedure for U_0 determination consists of stretching, performed at temperatures of the corresponding interrelaxation interval, at loading rates and clamping length ensuring the linearity of tension diagrams and destruction in 100–400 s. In this case parameter U_0 is determined by the formula:

$$U_0 = T_0 R \ln(\tau_{eff}/\tau_0),$$

where T_0 is the temperature determined by linear extrapolation of the temperature dependence of the ultimate tensile stress $\sigma(T)$ within an inter relaxation interval, $\sigma = 0$; τ_0 is a constant equal to 10^{-12} s for rubbers; τ_{eff} is the effective durability at indicated test conditions on a tensile-testing machine, calculated by the formula:

$$\tau_{eff} = 3.7 \cdot 10^{-2} \frac{T_m}{T_0 - T_m} t$$

where T_m is the temperature of an interval middle within which U_0 is determined; t is time of a sample deformation before rupture.

To demonstrate possibilities of the new method the rubber durability was determined on the basis of ethylene propylene rubber in a wide temperature range, including higher temperatures at which the GOST methods do not work. The tests were performed on a yMUB-4 device. The samples had the form of a double blade with a 10 × 1 × 1 m operating part, tension rate of 5 mm/min; heating time of 8 min; temperature range of 20–300°C.

Table I presents the initial data to calculate U_0, as well as the computed values of U_0 and τ corresponding to three temperature intervals.

In every temperature interval the relative elongation at rupture, ε_p, was virtually constant for all temperatures within an interval, indicating a stable structure inside them. The transition from low to higher temperature intervals is accompanied by

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TABLE I

Calculation of rubber durability based on the ethylene-propylene raw rubber

Stage	Temperature range, °C	Data for calculation	U_0 kJ/mole, τ
I	20–60	T_0 = 86°C ε_p = 80% t = 96 s τ_{eff} = 22 s	U_0 = 90, $\tau_{25°}$ = 125 years
II	60–140	T_0 = 246°C ε_p = 20% t = 24 s τ_{eff} = 2.2 s	U_0 = 122, $\tau_{140°}$ = 5.1 days
III	250–300	T_0 = 300°C ε_p = 15% t = 18 s τ_{eff} = 10 s	U_0 = 143, $\tau_{300°}$ = 8.7 sec

TABLE II

Variation of rubber properties under ionizing irradiation[2]

	Initial			After irradiation		
Sample	σ, MPa	U_0, kJ/mole	τ,[a] years	σ, MPa	U_0, kJ/mole	τ,[a] years
NBR-18SM	30	109	1,894	14.2	102	677
Nairite-M	75	121	11,033	22.2	105	1,063

[a]Durability correspond to 25°C.

an increase in U_0 which can be explained by partial linkage (formation of a three-dimensional net) in rubber.

This is also evidenced by a drastic decrease in ε_p from 80% to 15%. Comparison of the U_0 values obtained by the GOST method on the thermal ageing data (70, 90, 110, 195 and 150°C) and by the new procedure within the II temperature range (60–140°C) shows a good convergence of results: 126 and 122 kJ/mole, respectively. The determination of U_0 by the GOST method within the I and III intervals is impossible. Therefore, the calculation here was performed by the new procedure. The $\tau_{300°}$ value obtained corresponds to the results of bench tests.

In addition it is possible to estimate the contribution of ionizing radiation to the decrease in durability. This has been shown using raw rubbers made of butadiene-nitrile (NBR-18SM) and chlorprene (nairite M). To this end one-half of the samples of the two rubbers was ^{60}Co-γ irradiated at a dose of 50 Mrad (500 kgRa), and a dose rate of 400 rad/s (4 gRa/s). Table II shows that the ultimate tensile stress of NBR-18SM samples under irradiation decreases 2.1 fold and that of nairite-M samples, 3.4 fold, while parameter U_0, is decreased by 7 and 16 kJ/mole, respectively.

The effect of γ-radiation on the durability of rubbers of different polymeric types is inadequate: τ of irradiated NBR 18SM samples amounts to 35% of the initial samples while for nairite-M altogether 9.6%. At the same time, the durability of

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both rubbers after 500 kgRa irradiation is sufficiently high to satisfy specifications for rubbers requiring different technical objectives.[2]

Thus, the results of the present study show that the procedure developed has a number of advantages over the known methods used for estimation of the durability of polymeric materials.

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