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EFFECT OF PHENOL-FORMALDEHYDE RESIN ON THE RESISTANCE OF HIGH-PRESSURE POLYETHYLENE TO PROLONGED EXPOSURE TO ELECTRICAL DISCHARGES

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Abstract. The widespread use of polymer films necessitates a comprehensive investigation of their electrophysical and mechanical properties, as well as the changes in these properties under the influence of external factors during operation. The effect of prolonged exposure to electrical discharges on the mechanical and electrical strength, as well as on the structure of the polymer composition developed by us (HPPE + 0.05 wt% PR), has been studied. Experimental results show that the inclusion of 0.05 wt% phenol–formaldehyde resin (PR) is optimal and provides greater resistance to the effects of electrical discharges compared to both the original HPPE and HPPE containing other additive concentrations. The influence of phenol-formaldehyde resin in an optimal amount (0.05 wt%) on changes in electrical and mechanical strength under prolonged UV radiation in air was investigated. Using IR spectrophotometry, the contribution of the additive to the stability of the electrophysical properties of HPPE during ionization aging under the influence of electrical discharges has been established. It was found that in the modified HPPE film sample, the optical density of IR absorption bands corresponding to terminal carbonyl groups is consistently lower than that of the corresponding bands in the unmodified sample.

Keywords: High-pressure polyethylene, phenol-formaldehyde resin, mechanical strength, electrical strength, UV irradiation, IR spectrum, aging time.

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1 Introduction

The development of high-pressure polyethylene (HPPE) polymer compositions containing very low concentrations (0.01-0.1%) of nano- and micro-additives in polymer films - exhibiting enhanced electrophysical and mechanical properties, heat resistance, and aging stability during operation - was first undertaken in our research laboratory over a decade ago. The industrial implementation of these advanced nano- and micro-compositions could provide significant economic advantages.

In the technology of producing new polymer composite materials with improved physical-mechanical, electrical, and technological properties, it is essential to understand the influence of additives and external factors on the molecular-structural transformations occurring in polymers (Čubrić et al., 2022). Under the conditions of ionization processes in gas inclusions, polymers undergo electrical aging, which gradually alters their structure and deteriorates their electrophysical and mechanical characteristics (Conner et al., 1998; Ivanov & Solina, 2019; Zakrevsky et al., 2019).

Because of their high performance, low weight, cost-effectiveness, and ease of processing, polymer composites are widely used as structural and insulating materials, sealants, thermal insulators, and reinforcing agents. Currently, polyolefin-based composites find applications not only in aerospace and shipbuilding, but also in mechanical engineering, energy, electronics, electrical and radio engineering, transportation, construction, and other industrial sectors (Mehrabova et al., 2024a; Mahat et al., 2016; Steffen et al., 2010; Rajab et al., 2019).

Polyethylene-based materials exhibit high strength, resistance to aggressive environments and radiation, and excellent mechanical and dielectric properties. Moreover, polyethylene can be processed using all known methods for thermoplastics (Godzhaev et al., 2022; Zeynalov et al., 2023).

It is well established that exposure of polymers to partial electrical discharges in air at room temperature leads to oxidation, macromolecular degradation and cross-linking, an increase in dielectric losses, and a reduction in breakdown voltage, electrical and mechanical strength, as well as surface erosion of the polymer.

According to Mehrabova et al. (2025a), the introduction of a small amount of phenol–formaldehyde resin (0.05%) significantly enhances the mechanical strength of HPPE. In the present study, the mechanical durability of HPPE and its modified form was investigated at various temperatures, revealing an increase in resistance to the combined effects of external physical factors.

Furthermore, Slutsker (2008) reported that the electrical strength of HPPE film and its optimal modification (HPPE + 0.05 wt% PR) increases notably.

Polyolefin-based composites are promising materials for use as dielectrics in the electrical engineering industry due to their high performance characteristics, light weight, low cost, and ease of processing. However, the use of polyolefins (polyethylene and polypropylene) without additional modification methods, such as enhancing their electromechanical properties, ensuring stability under irradiation, resistance to electrical aging and mechanical loading, and improving processing technology often fails to meet modern operational requirements.

The aim of the present work is to study the electrical and mechanical properties of the HPPE-based polymer composition containing an optimal amount of phenol–formaldehyde resin (PR), with particular attention to its resistance to prolonged exposure to electrical discharges.

2 Methodology

The object of the study was high-density polyethylene (HPPE) of grade 10803-020, and the additive used was phenol–formaldehyde resin (PR) with the chemical formula $[-C_6H_3(OH) - -CH_2]_m$. The resin is characterized by mechanical stability, high strength, corrosion resistance, and excellent electrical insulating properties.

The additives were introduced into the HPPE raw material by mechanical mixing in quantities of 0.01–0.1 wt%. Prior to the addition of the additive, the HPPE was dispersed using sieve analysis on a particle size determination apparatus.

The sieve analysis setup consisted of a vibration drive and a set of sieves, typically comprising 5-6 sieves covering the full particle size range of the bulk material being tested. A sieve pan was positioned at the bottom to collect the finest fraction. The vibration drive imparted vibratory and rocking motion to the sieves. The sieving duration was 10 minutes for coarse-grained materials and about 20 minutes for fine-grained ones. The vibration time and intensity were controlled by a timer on the drive. The resulting particle sizes were less than 50 μm.

Polymer films were produced by the hot-pressing method using blanks obtained from an injection molding machine. A manual electrically heated hydraulic press (model PG-60) was used. The calculated amount of micro-additive was placed into a polished flat mold fitted with a 0.1 mm thick spacer. The mold was heated to 140–150 °C, and once the desired temperature was reached, the sample was compressed under a pressure of 150 atm. After cooling, the film

was removed from the mold.

Samples for measuring physical–mechanical, electrical, and thermal properties were prepared from the resulting films, which had a thickness of 50-60 μm. Film uniformity was evaluated by measuring thickness across the entire surface using both an HZB-2 optical thickness gauge and a micrometer. The thickness value for each sample was taken as the arithmetic mean of 10 measurements. Results from repeated experimental investigations of the mechanical, dielectric, optical, and structural properties confirmed uniform dispersion of the phenol-formaldehyde resin within the HPPE matrix.

The exposure of polymer dielectrics to electrical discharges was conducted in an asymmetrical test cell. To ensure a uniform, long-term electrical discharge without causing breakdown of the test specimen, a second dielectric (barrier) made of the same material or glass is often used in an asymmetrical cell.

This cell consisted of a system of metal electrodes between which the polymer film under study was placed. The lower electrode was a smooth stainless-steel plate measuring 180 × 130 mm, with one side nickel-plated. The polymer film, both before and after pre-stretching, was tightly mounted on this plate, which served as the grounded electrode. To maintain a constant air gap of 1.5 mm thickness between the upper electrode and the film, glass spacers were placed along the edges. A schematic diagram of this cell is shown in Figure 1. High-voltage, industrial-frequency power was supplied to the electrodes from the AII-70 apparatus.

The discharges were characterized by partial discharges in air (low-power localized electrical breakdowns). Tests were conducted at room temperature of 23-25^{0}C and ambient humidity of 40-50%.

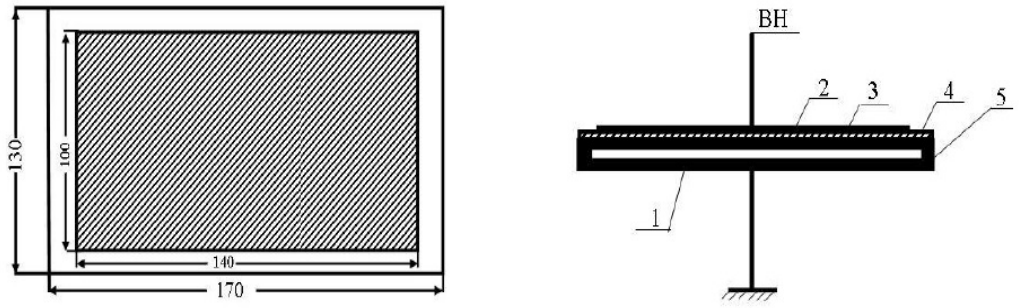

Figure 1: Test cell with an air gap: 1 – metal grounded electrode, 2 – glass metallized plate, 3 – metal coating, 4 – glass spacers, 5 – polymer film

The measurements of the specific volumetric electrical resistance (ρ_v) of the samples were carried out using an E6-13A teraohmmeter in the temperature range of 300–500 K, under linear heating at a rate of 3 deg/min, with errors ranging from ±2.5% to ±5%, depending on the specific measurement range. The dielectric permittivity (ε) and the dielectric loss tangent (tan δ) were measured using an R589 AC bridge at a frequency of 1 kHz over the temperature range of 300–500 K. The error in determining the dielectric permittivity (ε) is approximately ±0.2% of the measured value, while for the dielectric loss tangent (tan δ) it is ±(0.002 + 0.005%), depending on the specific measurement range. It should be noted that for each measurement, 8–10 samples ($n = 8-10$) were tested at a temperature of 23^{0}C and relative humidity of 40–50%.

An infrared (IR) spectrophotometer (Fig. 2) was used to record the absorption, transmission, and reflection spectra of the samples in the infrared region, as well as to perform qualitative and quantitative analyses of their composition.

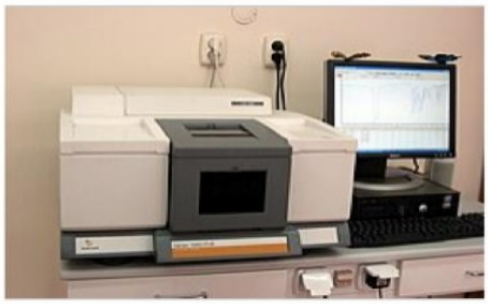

Figure 2: Research IR spectrometer

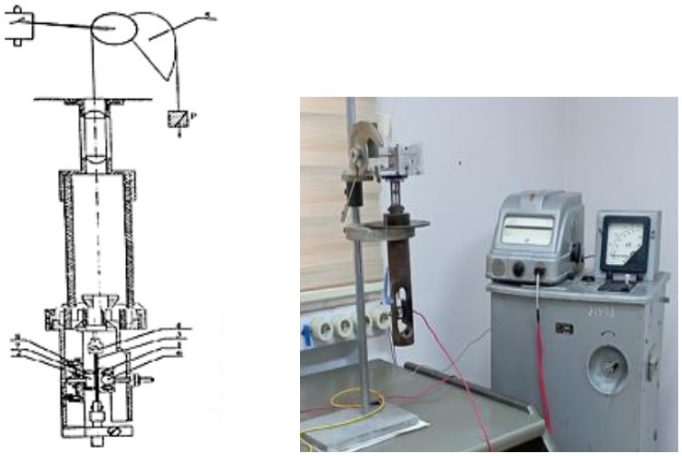

Figure 3: Setup for determining the electrical and mechanical strength of polymer film materials: 1, 2 – electrodes; 3 – sample; 4 – clamps; 5 – lever mechanism; 6 – ball; 7 – protective ring; 8 – springs

For mechanical strength testing, the samples were prepared in the form of double-strip specimens measuring 20 mm in length and 2 mm in width. Tests on the time dependence of mechanical strength at room temperature (22 °C) were conducted using a tensile testing machine designed for small-sized specimens, which allows the application and maintenance of a constant tensile stress until sample failure (Fig. 3).

The test was performed at a crosshead speed of 1.2.5 mm/min, with an allowable deviation of ±0.3–0.4 mm/min. The values of σ and τ were determined at a temperature of 22 °C as the arithmetic mean of 8–10 independent measurements. $s = \frac{F}{S}$, where F is the applied force and S is the initial cross-sectional area. When calculating the values of σ_{st} and E_{st}, the two highest and the two lowest results were discarded.

3 Experimental Results

Figure 4 shows the time dependence of mechanical strength (the relationship between lg τ and σ) for original HPPE and its optimal modification (HPPE + 0.05 wt% phenol–formaldehyde resin) subjected to electrical discharges. The tests were conducted under conditions where $U = 9kV$ represents the electric voltage applied to the cell, and $t = 5h$ and $t = 19h$ denote the elapsed time from the moment of loading to sample failure under constant mechanical stress. As can be seen, the well-known linear relationship between lg τ and σ at constant temperature is observed, i.e., the dependence follows the empirical equation:

$$\tau = Aexp^{-a\sigma},$$

where A and α are constants depending on the material properties and the measurement temperature. From Fig. 4, it can be seen that all lines in the coordinates $\lg \tau$ on σ intersect the ordinate axis at the same point, the value of which corresponds to the parameter $lg\ \ A$. However, the slope of these lines, i.e., the coefficient α, increases with increasing t_{ag}.

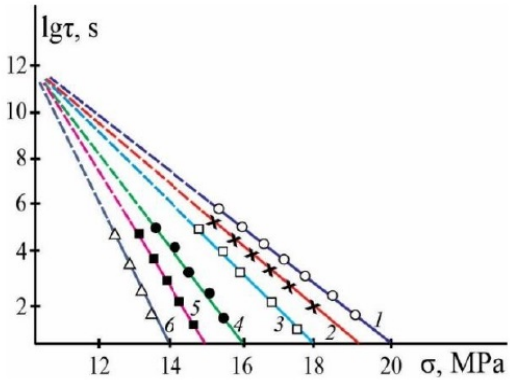

Figure 4: Time dependence of mechanical strength of HPPE and its optimal modification under exposure to electrical discharges in air: 1 – HPPE + 0.05 wt% PR (t = 0); 2 – HPPE + 0.05 wt% PR (t = 5 h); 3 – HPPE + 0.05 wt% PR (t = 10 h); 4 – HPPE (original) (t = 0); 5 – HPPE (original) (t = 5 h); 6 – HPPE (original) (t=10 h), U=9 kV

References Sokolova et al. (2022); Mehrabova et al. (2024b); Mammadov et al. (2024) indicate that the physical meaning of the parameter α lies in its characterization of the volume within which the elementary act of converting potential mechanical energy into thermal energy takes place. According to the given relationship, the fracture (destruction) process occurs as a result of fluctuations in the thermal motion energy. The applied stress merely facilitates this process by lowering the potential energy barrier between the pre-failure and post-failure states.

If a plot of mechanical strength versus aging time is constructed based on the data presented in Figure 4, then, as shown in Figure 5, a slight decrease in the mechanical strength of the polymer composition (HPPE + 0.05 wt% PR) is observed.

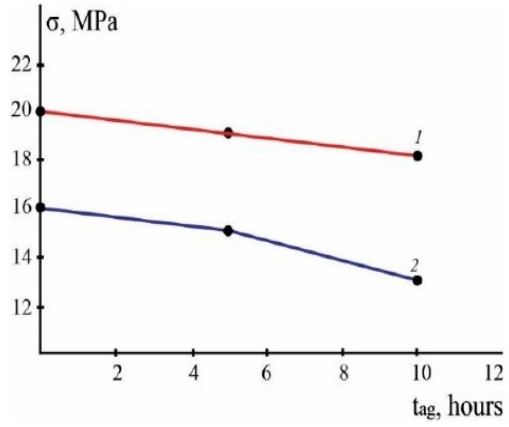

Figure 5: Dependence of mechanical strength of HPPE and its optimal modification on the duration of exposure to electrical discharges in air for HPPE + 0.05 wt% PR (1) and HPPE (without additive) (2).

Figure 6 shows the change in electrical breakdown strength of HPPE (without additive) and its optimal modification (HPPE + 0.05 wt% PR) as a function of aging time.

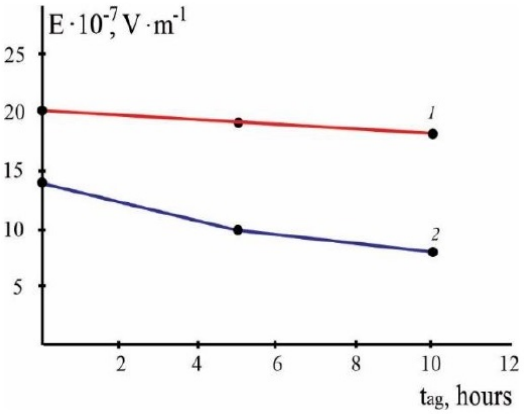

Figure 6: Dependence of electrical breakdown strength on the duration of exposure to electrical discharges in air for HPPE + 0.05 wt% PR (1) and HPPE (without additive) (1).

In both cases, a monotonic decrease in electrical strength is observed as a function of aging time. From Fig. 6, it is evident that the electrical strength of the HPPE + 0.05 wt% PR composition, after electrical aging, always remains higher than that of HPPE (without additive).

A good correlation is observed when studying the dependence of specific volume electrical resistivity (ρ_v), dielectric loss tangent ($tg\delta$) and dielectric permittivity (ε) of HPPE and its optimal composition HPPE + 0.05 wt% PR on the duration of exposure to electrical discharges in air, as shown in the table 1.

Table 1: Dependence of dielectric properties of HPPE and its optimal composition on the duration of exposure to electrical discharges.

Materials	**Properties**				
	t_{ag}, hour	E_{st}, $E \cdot 10^{-7}, V \cdot m^{-1}$	$tg\delta$ $f = 1$kHz, $t = 20^0$C	$\rho_v(Ohm \cdot m)$ $t = 20^0$ C	ε
HPPE + 0,05 wt % PR	0	20	$3 \cdot 10^{-4}$	$1 \cdot 10^{16}$	2,3
	5	19	$3,5 \cdot 10^{-4}$	$1 \cdot 10^{15}$	2,4
	10	18	$4,2 \cdot 10^{-4}$	$1 \cdot 10^{13}$	2,5
HPPE (initial)	0	14	$4,5 \cdot 10^{-4}$	$1 \cdot 10^{15}$	2,4
	5	10	$5 \cdot 10^{-4}$	$1 \cdot 10^{14}$	2,5
	10	8	$5,5 \cdot 10^{-4}$	$1 \cdot 10^{12}$	2,6

From the table 1, it can be seen that (ρ_v), ($tg\delta$) and (ε) of HPPE are also sensitive to the addition of phenol-formaldehyde resin, with a content of 0.05 wt% again proving to be optimal, as it provides the greatest stability of dielectric properties during electrical aging under the influence of discharges.

It is known that the effect of ionizing radiation on polyolefins is accompanied by polymer oxidation caused by reactions of free radicals with atmospheric oxygen. This oxidation of macromolecules largely determines the efficiency of their radiation-induced crosslinking and degradation. The addition of a small amount of PR (0.05 wt%) acts as a physical crosslinking agent for HPPE, increasing the packing density of polymer chains and thereby hindering the diffusion of ozone molecules into the HPPE, which in turn weakens oxidation processes.

Based on this, we investigated the effect of adding phenol-formaldehyde resin in an optimal amount (0.05 wt%) on changes in electrical and mechanical strength under prolonged UV exposure in air. UV irradiation was provided by a DRSh-500 lamp.

The table 2. shows the changes in the electrical and mechanical properties of HPPE (original) and its optimized modification (HPPE + 0.05 wt% PR) as a function of UV irradiation time (t_{rad}= 30–50 hours).

Table 2: Electrical and Mechanical Strength of HPPE and Its Optimal Composite vs. UV irradiation Time

Materials			
	t_{rad}, hours	σ_{st}, MPa	E_{st}, $E \cdot 10^{-7}$, $V \cdot m^{-1}$
HPPE + 0.05wt% PR	0	20	20
	30	19	20
	50	18	19
HPPE (original)	0	16	14
	30	14	11
	50	10	8

As shown in the table 2, the observed increase in the electrical and mechanical strength of HPPE remains stable after 30 hours of UV irradiation, with only a slight decrease following 50 hours.

Thus, the observed enhancement in strength and stability of the HPPE modification can be attributed to the structuring effect of the additive, which promotes tight packing of the macromolecules during the formation of the film.

It is well established that physicochemical changes occurring in polymer films under the influence of external factors—in this case, electrical discharges—are always accompanied by and, to some extent, correlated with structural transformations. As shown in Figures 4 and 5, at a constant stress level, the mechanical strength of both HPPE films and their optimal modification decreases with increasing exposure time.

This reduction in mechanical strength can be attributed to two primary factors (Dattelbaum et al., 2020; Mehrabova et al., 2025b): an increase in brittleness of the film caused by the formation of dense cross-links, and degradation processes induced by prolonged exposure to electrical discharges.

In the present case, the decrease in mechanical strength of the films appears to be predominantly associated with degradation processes (Mehrabova et al., 2025b; Mohammad & Fatemi , 2020; Li et al., 2018), as the presence of oxygen accelerates the degradation reaction. The slight reduction in the mechanical and electrical strength of HPPE films aged with a small addition of phenol–formaldehyde resin (PR) also indicates the formation of densely packed structures with a fine spherulitic organization. Such a structure impedes oxygen diffusion and thereby suppresses ionization processes during mechanical failure. This observation is in good agreement with the changes observed in the IR spectra.

Therefore, structural changes that occur after exposure to electrical discharges in HPPE and HPPE + 0.05 wt% PR were examined using infrared (IR) spectrophotometry. The absorption spectra revealed characteristic bands associated with oxygen-containing functional groups.

Figure 7 presents the dependence of the optical density of the carbonyl group absorption bands for HPPE and its optimal modification on the duration of exposure to electrical discharges. It can be seen that with increasing exposure time in air, a linear decrease in the optical density of the carbonyl group bands is observed.

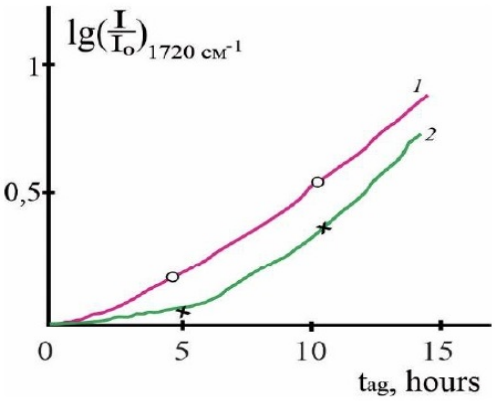

Figure 7: Dependence of the optical density of absorption bands of carbonyl groups in HPPE film and its optimal modifications on the duration of exposure to electrical discharges in air: 1 – HPPE (original), 2 – HPPE + 0.05 wt% PR.

It is also evident from Figure 7 that, in all cases, the intensity of the carbonyl group bands increases with the duration of electrical aging. However, across all exposure conditions, the optical density of the absorption bands in HPPE films containing the phenol-formaldehyde resin (PR) additive at the optimal concentration remains lower than that of the corresponding bands in the unmodified HPPE films.

Since the optical density of IR absorption is approximately proportional to the concentration of the corresponding functional groups in the sample, it can be concluded that the addition of phenol-formaldehyde resin effectively reduces chemical transformations in HPPE caused by oxidation, degradation, and cross-linking of polymer chains.

This finding further confirms that a small amount of PR (0.05 wt%) acts as a physical structure-forming agent within the HPPE matrix, increasing the packing density of the polymer chains and thereby hindering the diffusion of ozone molecules into the polymer. As a result, the oxidation processes are significantly suppressed, enhancing the structural and electrophysical stability of the material.

4 Conclusion

In this study, a high-pressure polyethylene (HPPE)–based composition was developed, characterized by the incorporation of a very small amount (0.05 wt%) of phenol-formaldehyde resin (PR) as an organic additive with excellent technological compatibility. The influence of electrical discharges on the mechanical strength, electrical strength, and structural characteristics of HPPE and its optimal composition was investigated as a function of exposure time (t).

It was established that, with increasing exposure time to electrical discharges at a constant applied voltage (U), the mechanical strength of both HPPE and its optimal modification decreases monotonically during aging in air. However, the addition of a small amount of PR (HPPE + 0.05 wt% PR) notably reduces the rate of mechanical degradation, particularly at relatively low voltage values, indicating a stabilizing effect of the additive.

The influence of phenol-formaldehyde resin in an optimal amount (0.05 wt%) on changes in electrical and mechanical strength under prolonged UV radiation in air was investigated. The observed increase in the electrical and mechanical strength of HPPE remains unchanged after UV irradiation, ensuring their stability during irradiation.

Furthermore, the optimal HPPE composition exhibits enhanced resistance to electrical discharges, as confirmed by the reduced intensity of the C=O absorption bands in its IR spectra. Under identical experimental conditions and with the same additive concentration, the optical

density corresponding to the terminal carbonyl group absorption bands in the modified HPPE films is significantly lower than in the unmodified material.

These results demonstrate that the introduction of 0.05 wt% phenol–formaldehyde resin effectively suppresses oxidation and degradation processes, improves the structural stability, and enhances the electrical and mechanical durability of HPPE under prolonged exposure to electrical discharges.

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