Inhibition of Polyethylene and Polypropylene Oxidation by Nitro Compounds Reacting with Alkyl and Peroxy Radicals

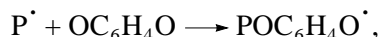## Yu. B. Shilov and E. T. Denisov

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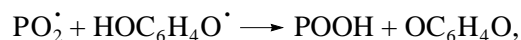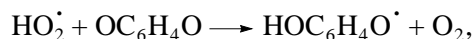Received November 25, 1999

Abstract—The kinetics of chain termination in polypropylene and polyethylene oxidation inhibited by 2,4-dinitrololuene is studied. 2,4-Dinitroluene inhibits the oxidation of polyethylene and polypropylene lacking hydroperoxide groups by terminating chains in the reaction with alkyl macroradicals. In oxidized polypropylene containing hydroperoxide groups, chain termination on 2,4-dinitrololuene involves both alkyl and peroxy radicals. A cyclic mechanism is proposed for chain termination in partially oxidized polypropylene involving $HO_2^{\bullet}$ and nitroxyl radicals formed from 2,4-dinitrololuene.

INTRODUCTION

Earlier, we found that quinones retard the oxidation of a polymer (PH) by terminating chains in the reaction with alkyl macroradicals [1, 2]. In oxidized polypropylene, their action is more efficient because chains are terminated also in the reaction with $HO_2^{\bullet}$ radicals. The latter are generated from hydrogen peroxide, which, in turn, is formed from hydroperoxide groups of a polymer (POOH) [3–5]. This complex inhibition by quinones can be due to their ability to add alkyl radicals in the reaction with a polymer macroradical $P^{\bullet}$

$$P^{\bullet} + OC_6H_4O \longrightarrow POC_6H_4O^{\bullet},$$

on the one hand, and to participate in the redox cycle

$$HO_2^{\bullet} + OC_6H_4O \longrightarrow HOC_6H_4O^{\bullet} + O_2,$$

$$PO_2^{\bullet} + HOC_6H_4O^{\bullet} \longrightarrow POOH + OC_6H_4O,$$

on the other. Stable nitroxyl radicals exhibit the same properties. They break chains in the reaction with alkyl radicals [6, 7] and react with the $HO_2^{\bullet}$ hydroperoxy radicals in oxidized polypropylene [2, 8].

Nitro compounds are obviously similar to quinones: they can add alkyl radicals and act as oxidants. Moreover, they can produce nitroxyl radicals. Therefore, we expected that nitro compounds like quinones retard polymer oxidation by breaking chains in the reactions with both alkyl and peroxy (hydroperoxy) radicals. Our preliminary experiments confirmed this assumption. This work is devoted to the comprehensive kinetic study of the oxidation of two polymers, polypropylene and polyethylene, inhibited by 2,4-dinitrotoluene.

EXPERIMENTAL

Reagents. We chose for the study isotactic polypropylene (a commercial specimen purchased from the Moscow Oil Refining Plant). Its oxidation kinetics was studied in detail in [9]. Polypropylene purified as described in [9] was used as a finely divided powder (molecular weight, 2.8×10^5 g/mol; ash content, 0.02%; the fraction soluble in boiling *n*-heptane, 2.5%). Polyethylene was prepared by ethylene polymerization on a titanocene catalyst in ethyl chloride.[1] Polyethylene with less than five branchings per 1000 monomeric links was characterized by a specific viscosity [η] of 0.19 dl/g (decalin, 408 K) and the ratio $M_w/M_n = 2.5$ (where M_w and M_n are the average weight and average molecular weight, respectively). 2,4-Dinitrotoluene (reagent grade) was doubly recrystallized from ethanol. The inhibitor (2,4-dinitrololuene, DNT) and the initiator (cumyl peroxide, CP) were introduced into polyethylene and polypropylene from a benzene solution. Then, the resulting polymer (polyethylene or polypropylene) was dried at room temperature. The time required for the complete removal of the solvent from polypropylene was estimated from changes in the EPR spectrum of the stable nitroxyl radical (2,2,6,6-tetramethylpiperidine oxide). This estimation method implies that the solvent in the polymer acts as a plasticizer and causes the rapid rotation of the nitroxyl radical and, hence, a change in its EPR spectrum. The spectrum changes as the solvent is removed from the polymer and acquires a shape typical of a dry polymer upon complete solvent removal. The time necessary for the complete removal of the solvent ranged from 10 to 15 h.

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[1] We thank G.P. Belov for the preparation of the polyethylene specimen.

Experimental procedure. Polypropylene oxidation initiated by cumyl peroxide or the hydroperoxide groups of oxidized polypropylene (POOH) was conducted in the presence of 2,4-dinitrololuene at 388.8 ± 0.2 K. Polypropylene containing hydroperoxide groups was obtained by its oxidation at 370 K in the presence of benzoyl peroxide, which was extracted with a benzene–alcohol solution after polypropylene oxidation. Oxygen consumption during polypropylene oxidation was measured using a pressure gauge at $P_{O_2} = 745 \pm 15$ torr. The degree of polypropylene oxidation was at most 0.15 mol/kg. Polyethylene was oxidized according to the same procedure.

RESULTS AND DISCUSSION

Kinetic parameter estimation. The initiation rate w_i was calculated by the equation $w_i = k_i[I]_0$, where k_i is the rate constant for initiation, and I is the initiator. For cumyl peroxide in polypropylene at 388.8 K, $k_i = 4 \times 10^{-6}$ s^{-1} [10]. The initiation rate of oxidation by hydroperoxide groups POOH of partially oxidized polypropylene was determined from the rate (w_0) of oxygen consumption according to the formula $w_i = w_0^2/a^2$ [10]. The a coefficient (mol kg^{-1} s^{-1})$^{1/2}$ was determined in the experiments on polypropylene oxidation initiated by cumyl peroxide at a certain w_i value. For polypropylene oxidation at 388.8 K, the a coefficient was 3.4×10^{-2} (mol kg^{-1} s^{-1})$^{1/2}$. For polyethylene oxidation in the presence of cumyl peroxide (5.0×10^{-2} mol/kg, $w_i = 2 \times 10^{-7}$ mol kg^{-1} c^{-1}, 388.8 K), the POOH group concentration attained its maximal value (3×10^{-2} mol/kg) and remained unchanged during further polyethylene oxidation. The initiation rate during POOH decomposition in oxidized polyethylene at 388.8 K was measured using the method of complex initiation. The experiments were conducted at a variable cumyl peroxide concentration and [POOH] $\cong 3 \times 10^{-2}$ mol/kg. The k_i^{POOH} value was determined by the equation

$$w_0^2 = a^2 k_i^{POOH}[POOH] + a^2 k_i^{CP}[CP]. \qquad (1)$$

From the slope of the w^2-vs.-[CP] plot, we determined that the $a^2 k_i^{CP}$ value and then the a coefficient were equal to 0.017 (mol kg^{-1} s^{-1})$^{1/2}$. From the intercept on the ordinate axis ($a^2 k_i^{POOH}$ [POOH] = 4.8×10^{-11} (mol kg^{-1} s^{-1})^2), we obtained $k_i^{POOH} = 5.5 \times 10^{-6}$ s^{-1}.

The efficiency of the reaction inhibition by dinitrotoluene was characterized by parameter F (kg/mol) [4, 10]:

$$F = \frac{w_i(1 - w^2/w_0^2)}{w[DNT]}$$
$$= \frac{w_i \beta}{w[DNT]} = G + (D/P_{O_2}), \qquad (2)$$

where w and w_0 are the rates of oxygen consumption in the reaction with and without the inhibitor, respectively. The G and D parameters characterize the efficiency of chain termination in the reaction of the inhibitor with peroxide (PO$_2^{\cdot}$) and alkyl (P$^{\cdot}$) macroradicals, respectively. They are $G = f k_7/k_2[PH]$ and $D = f k_9/k_1 \gamma$, where k_1 and k_2 correspond to the P$^{\cdot}$ + O$_2$ and PO$_2^{\cdot}$ + PH reactions, respectively; k_7 and k_9 characterize the reactions of the inhibitor with the peroxy and alkyl radicals, respectively; γ is the Henry coefficient for O$_2$ in an amorphous polymer phase; and f is the stoichiometric inhibition coefficient. The retardation by the binary inhibitor system was expressed in terms of the sum of the F_1 and F_2 parameters that characterize the retardation by either of the two individual inhibitors. We have $F_\Sigma \cong F_1 + F_2$ for the additive action of the inhibitors taken in equivalent concentrations and $F_\Sigma < F_1 + F_2$ for their nonadditive action. This allowed us to compare the efficiencies of the individual inhibitors and their mixtures to break the oxidation chains.

Kinetics of inhibited oxidation. The experiments with 2,4-dinitrololuene showed that it inhibited the initiated oxidation of polypropylene (Fig. 1). 2,4-Dinitrotoluene is less efficient than a phenol inhibitor (irganox 1010). Figure 1 (curve *4*) shows that irganox 1010 (PhOH, 1.0×10^{-3} mol/kg) retards polypropylene oxidation much more efficiently than 2,4-dinitrololuene (2.32×10^{-2} mol/kg) (curve *3*). The experiments with different 2,4-dinitrololuene concentrations carried out at various partial oxygen pressures suggested that the rate of the initiated oxidation of polypropylene inhibited by 2,4-dinitrololuene can be described by Eq. (2) with $G = 0$ and $D \neq 0$ (see Fig. 2). This means that 2,4-dinitrololuene breaks the chains in oxidized polypropylene only in the reaction with alkyl radicals. Therefore, chain termination in this system occurs by the same pathway as in radical polymerization [11], that is, via the reactions

$$P^{\cdot} + RNO_2 \longrightarrow RN(O^{\cdot})OP,$$

$$P^{\cdot} + RN(O^{\cdot})OP \longrightarrow RN(OP)_2,$$
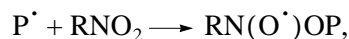
where RNO$_2$ is DNT.

Similar results ($F \sim 1/P_{O_2}$, see Fig. 2) were obtained in the experiments on the initiated oxidation of polyethylene with 2,4-dinitrololuene additives. We found the D
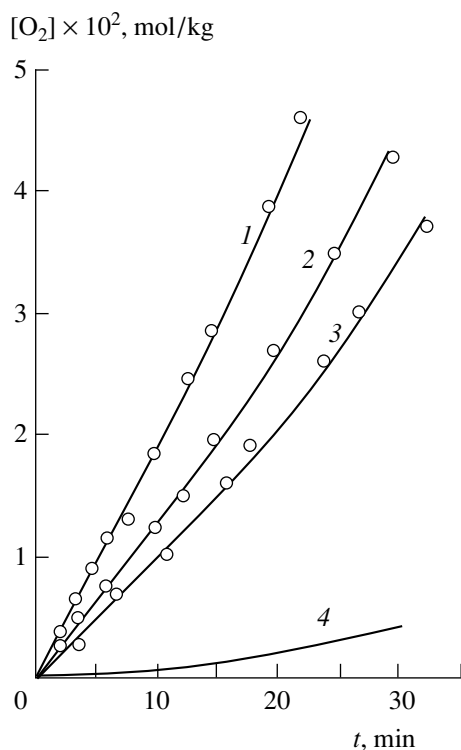
Fig. 1. Kinetics of oxygen consumption during polypropylene oxidation with cumyl peroxide ($w_i = 7.2 \times 10^{-7}$ mol kg^{-1} s^{-1}) at $T = 388.8$ K and $P_{O_2} = 745$ torr (*1*) without and (*2*) with the inhibitor, [DNT] $\times 10^2$, mol/kg: (*2*) 1.16 and (*3*) 2.32. (*4*) Oxygen consumption in the presence of irganox 1010 ([PhOH] $= 1.0 \times 10^{-3}$ mol/kg, $w_i = 4 \times 10^{-7}$ mol kg^{-1} s^{-1}, 388 K) [9].
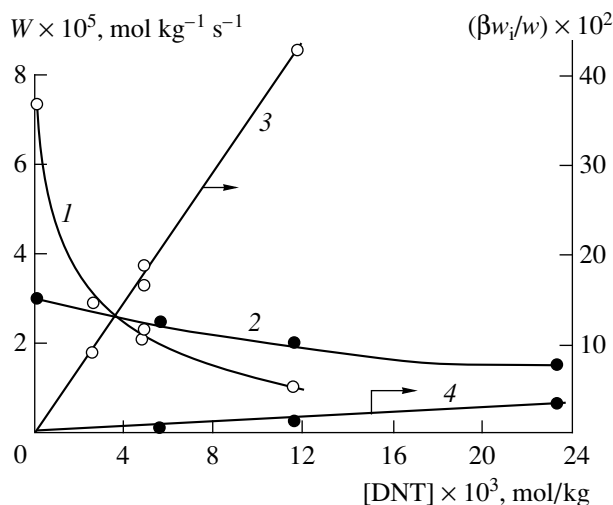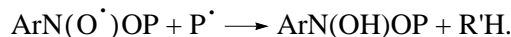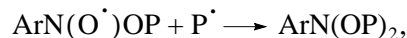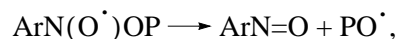

Fig. 2. A plot of (*1, 2*) the rate of polypropylene oxidation (*w*) and (*3, 4*) $\beta w_i/w$ ratio vs. [DNT] in the experiments with the initiation by (*1, 3*) the hydroperoxide groups ([POOH] = 10.86×10^{-2} mol/kg, $w_i = 4.6 \times 10^{-6}$ mol kg^{-1} s^{-1}) and (*2, 4*) CP ($w_i = 7.2 \times 10^{-7}$ mol kg^{-1} s^{-1}) at 388.8 K.

values equal to 1.5 ± 0.1 and 2.6 ± 0.5 kg atm mol^{-1} for polypropylene (389 K) and polyethylene (388 K), respectively.

Earlier studies on polypropylene oxidation inhibited by 2,6-dinitrophenol at 387 K showed that this inhibitor breaks chains both in the reactions with peroxy and alkyl radicals and that $F = 2.1 + 1.6/P_{O_2}$ (kg/mol). The first term, $G = 2.1$ kg/mol, characterizes chain termination in the reaction of the phenol group of the inhibitor with the $PO_2^{\cdot}$ radicals. The numerator of the second term, $D = 1.6$ kg atm mol^{-1}, is close to the D value of 1.5 kg atm mol^{-1} for dinitrotoluene and, obviously, characterizes chain termination in the reaction of alkyl radicals with precisely the nitro groups of dinitrophenol. One can easily see that the nitro groups in 2,4-dinitrotoluene and 2,6-dinitrophenol exhibit nearly the same activity when acting as alkyl radical acceptors.

The $ArN(O^{\cdot})OP$ nitroxyl radical in the case of this chain termination can undergo the following alternative transformations:

$$ArN(O^{\cdot})OP \longrightarrow ArN{=}O + PO^{\cdot},$$

$$ArN(O^{\cdot})OP + P^{\cdot} \longrightarrow ArN(OP)_2,$$

$$ArN(O^{\cdot})OP + P^{\cdot} \longrightarrow ArN(OH)OP + R'H.$$

Inhibition activity of the products of 2,4-dinitrololuene transformation. The inhibition activity of high-molecular products formed during polypropylene oxidation retarded by 2,4-dinitrololuene was estimated as follows. Polypropylene with 2,4-dinitrololuene (1.16×10^{-2} mol/kg) and cumyl peroxide as an initiator ($w_i = 7.2 \times 10^{-7}$ mol kg^{-1} s^{-1}) was stored in an inert atmosphere (N$_2$) at 388.8 K for 80 min. After that, low-molecular products were extracted with a benzene–alcohol solution from the polymer. The retardation of the oxidation of such polypropylene initiated by cumyl peroxide ($w_i = 7.1 \times 10^{-7}$ mol kg^{-1} s^{-1}) indicates the formation of high-molecular products of 2,4-dinitrololuene transformation and their participation in the inhibition of polypropylene oxidation. Assuming that the concentration of these products is $[P] \cong w_i\, t \cong 1.7 \times 10^{-3}$ mol/kg, the F_2 value of 10.6 kg/mol calculated for them is much higher than $F_1 = 1.5$ kg/mol for the initial nitro compound 2,4-dinitrololuene. The inhibition activity of the high-molecular products of 2,4-dinitrololuene transformation increases on passing from O$_2$ ($F_2 = 10.8$ kg/mol) to air ($F_2 \cong 17.3$ kg/mol) at $w_i \cong 1.1 \times 10^{-5}$ mol kg^{-1} s^{-1}. This fact suggests that the products of 2,4-dinitrololuene transformation can participate in chain termination in the reaction with the alkyl radicals of a polymer. If the system contains both a nitro compound (2,4-dinitrololuene) and high-molecular products, their joint retardation action becomes more pronounced and equal to

$F_\Sigma \cong 2.7$ kg/mol in terms of [DNT] = 1.16×10^{-2} mol/kg at $w_i = 7.2 \times 10^{-7}$ mol kg^{-1} s^{-1}. This F_Σ value is virtually identical to the value of $F_\Sigma = F_1 + F_2[P]/[DNT]$ expressed as a sum of the parameters $F_1 = 1.5$ kg/mol and $F_2 \cong 10.8$ kg/mol that characterize the retardation by each individual inhibitor acting in parallel.

Retardation by 2,4-dinitroluene with oxidized polypropylene. In oxidized polypropylene, containing hydroperoxide groups POOH, the efficiency of the nitro compound 2,4-dinitrololuene as an inhibitor of radical-chain oxidation of polypropylene markedly increases (Figs. 2 and 3, curves *1*). We obtained $F \cong 37.3 \pm 2.1$ kg/mol in oxidized polypropylene with [POOH] = 10.86×10^{-2} mol/kg at $P_{O_2} = 1$ atm (388.8 K) and various 2,4-dinitrololuene concentrations (Fig. 3, curve *1*) and $F = 1.5$ kg/mol in the initial polypropylene without POOH groups under similar conditions (Fig. 3, curve *4*). The efficiency of 2,4-dinitrololuene as an inhibitor of polypropylene oxidation in polypropylene containing hydroperoxide groups is 25 times higher than in nonoxidized polypropylene.

The manner in which the *F* parameter varies with the oxygen pressure suggests that 2,4-dinitrololuene in oxidized polypropylene with hydroperoxide groups breaks chains in the reactions with both alkyl and peroxy radicals: $G \cong 22$ kg/mol and $D \cong 18$ kg atm mol^{-1} (Fig. 3, curve *1*). The more significant retardation by 2,4-dinitrololuene is also observed when cumyl peroxide and the groups of oxidized polypropylene initiate oxidation simultaneously (Fig. 3, curve *2*). Note that the presence of CP in oxidized polypropylene containing POOH groups somewhat reduces the efficiency of retardation compared to the case when oxidation is initiated by only the POOH groups of oxidized polypropylene:

[ROOH], mol/kg	10.86×10^{-2}	6.94×10^{-2}
[CP], mol/kg	0	5.6×10^{-2}
G, kg/mol	0	20
D, kg atm mol^{-1}	18	≈ 2.0

The stoichiometric coefficient of the inhibition (*f*) of polypropylene oxidation by dinitrotoluene was estimated as follows. The introduction of 2,4-dinitrololuene (2.67×10^{-3} mol/kg) into oxidized polypropylene containing POOH groups (10.86×10^{-2} mol/kg, $w_i = 4.6 \times 10^{-6}$ mol kg^{-1} s^{-1}) in the air caused the complete inhibition of the process in ~40 min (Fig. 4, curve *4*), that is, no oxygen consumption occurred within this period of time. The POOH concentration after 40 min, just at the beginning of oxygen consumption, was [POOH] = 8×10^{-2} mol/kg. The inhibition coefficient *f* calculated from the equation $f = \Delta[POOH]_t e/[DNT]$ is ~3.6, where the radical escape into the bulk during POOH decomposition is $e = w_i/w_d \sim 0.33$, w_d is the rate of hydroperoxide group decomposition and $\Delta[POOH]_t$ is the hydroperoxide fraction decomposed in $t = 40$ min.
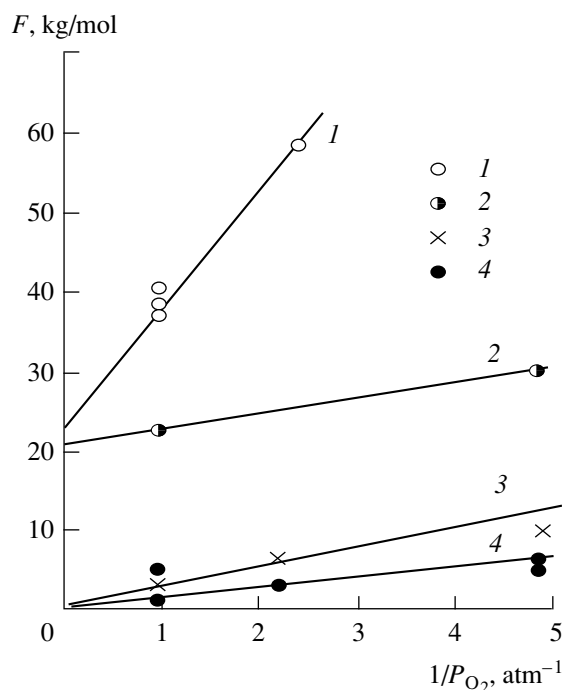

Fig. 3. A plot of *F* vs. $1/P_{O_2}$ for the oxidation of solid (*1*, *2*, *4*) polypropylene and (*3*) polyethylene in the presence of 2,4-dinitrololuene at 388.8 K: (*1*) initiation by the POOH groups: [POOH] = 10.86×10^{-2} mol/kg ($w_i = 4.6 \times 10^{-6}$ mol kg^{-1} s^{-1}), [DNT] $\times 10^3$, mol/kg: 11.62, 4.84, and 2.71 at $P_{O_2} = 1$ atm and 2.6×10^{-3} at $P_{O_2} = 0.4$ atm; (*2*) joint initiation by CP and the POOH groups: [POOH] = 6.94×10^{-2} mol/kg ($w_i = 2.2 \times 10^{-7}$ mol kg^{-1} s^{-1}), [CP] = 5.5×10^{-2} mol/kg ($w_i = 2.2 \times 10^{-7}$ mol kg^{-1} s^{-1}), [DNT] = 2.9×10^{-3} mol/kg; (*3*) initiation by CP (388.5 K, $w_i = 1.12 \times 10^{-6}$ mol kg^{-1} s^{-1}), [DNT] $\times 10^3$, mol/kg: 11.62 at $P_{O_2} = 1.0$ and 0.4 atm and 5.75 at $P_{O_2} = 0.21$ atm; (*4*) initiation by CP ($w_i = 7.2 \times 10^{-7}$ mol kg^{-1} s^{-1}): [DNT] $\times 10^3$, mol/kg: 11.62 and 5.81 at $P_{O_2} = 1$ atm, 11.62 at $P_{O_2} = 0.42$ atm, and 11.62 and 5.81 at $P_{O_2} = 0.21$ atm.

For comparison, we carried out similar experiments with oxidized polyethylene containing POOH groups (3×10^{-2} mol/kg) in the presence of 2,4-dinitrololuene at 388.8 K. The hydroperoxide groups in oxidized polyethylene are more stable than in polypropylene and decompose to radicals with a rate constant $k_i = 5.5 \times 10^{-6}$ s^{-1} ($w_i = 1.6 \times 10^{-7}$ mol kg^{-1} s^{-1}). Therefore, the oxidation of polyethylene containing POOH groups in the presence of 2,4-dinitrololuene was initiated by cumyl peroxide. We experimentally showed that the presence of POOH groups in oxidized polyethylene does not affect the antioxidant action of 2,4-dinitrololuene. This is the difference between polyethylene and polypropylene oxidations: chain termination in oxidized polypropylene containing the POOH groups occurs in the reac-
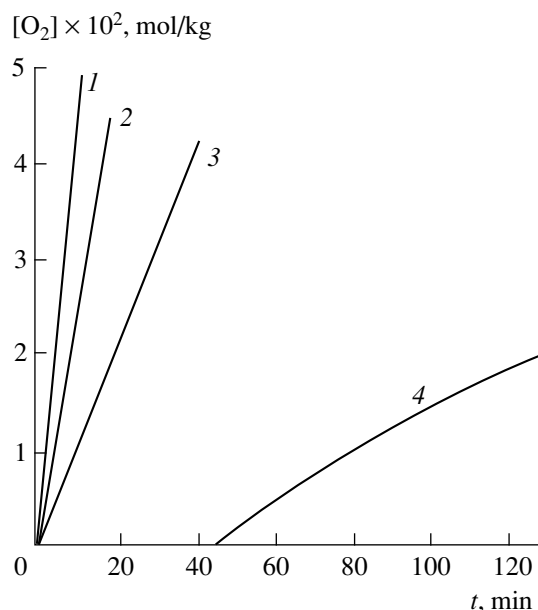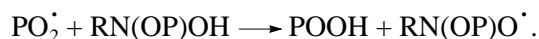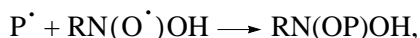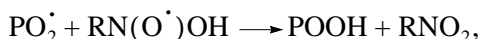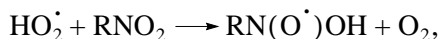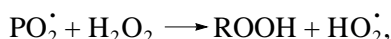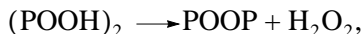
$[O_2] \times 10^2$, mol/kg

Fig. 4. A polypropylene oxidation (388.8 K) containing POOH (10.86×10^{-2} mol/kg): (*1*) without inhibitor, P_{O_2} = 1 atm; (*2*) without inhibitor in the air; (*3*) in the presence of 2,4-dinitrololuene (2.67×10^{-3} mol/kg), P_{O_2} = 1 atm; and (*4*) in the presence of 2,4-dinitrololuene (2.67×10^{-3} mol/kg) in the air.

tions with both alkyl and with peroxy radicals and, hence, is more intensive than that in nonoxidized polypropylene.

Mechanism of retardation by 2,4-dinitrololuene. More dramatic retardation by 2,4-dinitrololuene in oxidized polypropylene can be attributed to the formation of noticeable amounts (40–50%) of associated β-OOH groups during polypropylene oxidation. These groups decompose to H_2O_2, which is the source of $HO_2^{\bullet}$ radicals [4, 12, 13]. These radicals exhibit both oxidizing and reducing properties and, therefore, are responsible for chain termination in the reaction with 2,4-dinitrololuene. By analogy with the retardation action of quinones and nitroxyl radicals [14] in oxidized polypropylene, we propose the following mechanism for chain termination involving the nitro groups of 2,4-dinitrotoluene:

$$(POOH)_2 \longrightarrow POOP + H_2O_2,$$

$$PO_2^{\bullet} + H_2O_2 \longrightarrow ROOH + HO_2^{\bullet},$$

$$HO_2^{\bullet} + RNO_2 \longrightarrow RN(O^{\bullet})OH + O_2,$$

$$PO_2^{\bullet} + RN(O^{\bullet})OH \longrightarrow POOH + RNO_2,$$

$$HO_2^{\bullet} + RN(O^{\bullet})OH \longrightarrow H_2O + RNO + O_2,$$

$$P^{\bullet} + RN(O^{\bullet})OH \longrightarrow RN(OP)OH,$$

$$PO_2^{\bullet} + RN(OP)OH \longrightarrow POOH + RN(OP)O^{\bullet}.$$

This scheme implies that chain termination can occur via the cyclic mechanism.

CONCLUSION

We experimentally confirmed that dinitrotoluene retards polyethylene and polypropylene oxidations. The oxidation of nonoxidized polypropylene is inhibited in its reaction with alkyl macroradicals. The oxidation of both nonoxidized and oxidized polyethylene is also retarded only in the reaction of 2,4-dinitrololuene with alkyl macroradicals. The retardation mechanism principally changes upon 2,4-dinitrololuene introduction into oxidized polypropylene containing block hydroperoxide groups: the retardation efficiency of 2,4-dinitrololuene abruptly increases because of the intense chain termination in its reaction with peroxy radicals. This fact may be due to H_2O_2 formation from the block hydroperoxide groups followed by H_2O_2 decomposition to $HO_2^{\bullet}$ radicals and the rapid reaction of these radicals with 2,4-dinitrololuene. Chain termination can also occur via the cyclic mechanism under these conditions.

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