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Cavitation-Erosion Wear Resistance of Fluoroplastics in Model Food Production Media

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Abstract—The effect of acidic, alkaline and neutral media on wear kinetics and mechanism for fluoroplastics and polyolefins has been studied. Equations for fluoroplastic and polyolefin wear intensity are obtained based on the thermochemical and mechanical model of their destruction. The values of the activation energies for the surface destruction onset over the entire wear cycle of the polymer surface spherulite are determined.

Keywords: polymers, cavitation, fluoroplastics, corrosive media, corrosion-mechanical wear

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INTRODUCTION

Under corrosion-mechanical wear (CMW), whose subtype is represented by cavitation-erosion wear (CEW) of metals in corrosive media, the corrosion component of destruction is insignificant by itself. However, as it is a catalyst for electrochemical processes, it significantly affects the general characteristics of metal surface destruction [1]. The inertness of most polymeric materials, especially fluoroplastics, with respect to corrosive liquids has resulted in increased interest in the use of such materials for manufacturing products that contact chemically aggressive liquids.

This is also caused by the fact that fluoroplastics have good thermal and electrical insulation properties, they exhibit a significantly low density (three-fold lower than the density of metals and alloys, on average). The utilization coefficient for fluoroplastics is 0.89–0.95, whereas for metals it ranges within 0.5–0.6 [2]. In addition, the replacement of metals by polymers leads to a 5–6-fold reduction in labor input, as well as a 2–9-fold reduction in cost price [2].

Fluoroplastics are an extensive group of vinyl-type polymers, among which polytetrafluoroethylene (PTFE, fluoroplastic F4) and its modifications are the most widely used polymers in engineering. A distinctive feature of these polymers is their high crystallinity, reaching up to 85% [3]. It is recommended to use fluoroplastic products at a temperature not exceeding 250°C to exclude the possibility of a change in their structure and thereby mechanical characteristics. Studies on the heat resistance of fluoroplastics F4 and F4S15 according to Vika resulted in the values of

114 and 135°C, respectively. The results of the studies indicate that the mechanical properties of fluoroplastics significantly depend on temperature. With increasing temperature, the elastic modulus and the ultimate tensile strength exhibit a decrease, whereas the relative elongation increases [4].

As far as the chemical resistance is concerned, fluoroplastics surpass all known polymeric materials [2, 3]. Fluoroplastics do not dissolve in any of the known solvents, they are resistant with respect to acids and alkalis, they do not swell in water and cannot burn.

Superior mechanical chemical resistance characteristics inherent in fluoroplastics in comparison with other polymers have led to their widespread use in mechanical engineering as structural materials for the manufacture of elements operating in aggressive media (electrical insulation, antifriction, sealing elements in machine structures).

At the same time, the data concerning cavitation-erosion wear resistance for fluoroplastics are fragmentary and unsystematic. It is known from [4] that the cavitation-erosion wear resistance of fluoroplastic F4 is much higher than that of aluminum, but inferior to the wear resistance of alloyed steel, which in turn indicates that the use of polymers based on fluoroplastic F4 is promising for the manufacture of elements that are subjected to cavitation in aggressive media.

This work was aimed at studying cavitation-erosion wear resistance inherent in fluoroplastics in neutral, acidic and alkaline model media of food production.

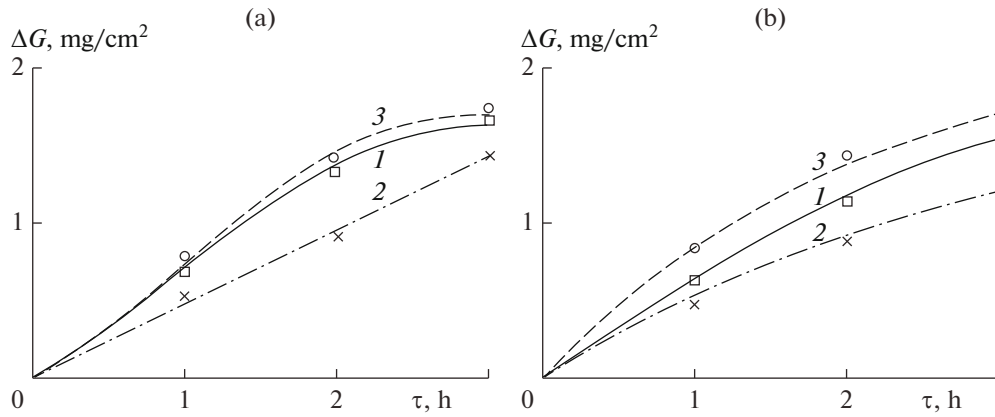


Fig. 1. Weight loss kinetics in the course cavitation-erosion wear: (a) fluoroplastic F4; (b) impact-resistant fluoroplastic F4S15. (1) neutral medium; (2) acidic medium; (3) alkaline medium.

MATERIALS AND METHODS

Cavitation-erosion wear resistance exhibited by fluoroplastic F4 and its impact-resistant analog F4S15 that contains 15% of fiberglass, has been studied in this work.

Our studies were carried out in model media. They were a neutral medium (3% solution of sodium chloride in distilled water), an alkaline medium (250 g/L of calcium oxide CaO + sucrose $\text{C}_{12}\text{H}_{22}\text{O}_{11}$, 15% by weight with respect to CaO), as well as an acidic medium (10 g/L of dibasic sodium phosphate Na_2HPO_4 + 5 g/L of citric acid $\text{C}_6\text{H}_8\text{O}_7$).

Testing for the cavitation-erosion wear resistance of the above-mentioned polymers was carried out using a specially designed and manufactured unit consisting of a UZDN-A ultrasonic generator, a sample holder installed in a tank for working media, wherein two helical-coil heat exchangers with oppositely directed coils were mounted. The heat exchangers provided temperature control in the entire volume of the medium in the tank and directly in the cavitation zone accurate within $\pm 2^\circ\text{C}$ throughout the entire period of the experiments. The gap between the ends of the vibrator and the samples was 0.6 mm, which provided the maximum rate of sample destruction and, accordingly, a decrease in the test duration time.

The oscillation amplitude of the vibrator was $a = 50 \mu\text{m}$, the frequency was $\lambda = 22 \text{ kHz}$ and the radiation power was $p = 150 \text{ W}$. The design of the unit makes it possible to avoid any influence of the temperature factor on the destruction of polymeric surfaces. The design features of the unit and the system for surface temperature stabilization are described in more detail in [5].

The photographic and video imaging of surface destruction for polymeric samples after and in the course of cavitation was carried out using a MIM-10 microscope and a Ricoh Caplio r7 digital camera [6].

RESULTS AND DISCUSSION

The analysis weight loss ΔG depending on the test time τ (Fig. 1) has shown that the lowest weight loss values for fluoroplastics are observed in an acidic medium, whereas the greatest values of weight loss are exhibited in an alkaline medium. At the same time, for fluoroplastic F4, the weight loss kinetics in an acidic medium is described by a linear dependence, and with increasing test time the weight loss exhibits an increase. In neutral and alkaline media, the ΔG – τ dependences have a decaying character (Fig. 1a).

The weight loss for impact-resistant fluoroplastic F4S15 is slightly lower in comparison with fluoroplastic F4 in all the media (Fig. 1b), and the ΔG – τ curves have an exponential shape, that is, the wear rate decreases with the course of the test duration time.

The identical shape of weight loss curves for F4S15 fluoroplastic (Fig. 1b) allows us to assume that the mechanism of its destruction in all the media does not change, whereas for fluoroplastic F4 in an acidic medium (Fig. 1a, line 2), the destruction mechanism is somewhat different from that in neutral and alkaline media.

The visual assessment of the wear for the surface of the studied polymers shows their identical destruction character [7]. The cavitation-erosion wear begins in all the media at the grain boundaries of polymers or from the amorphous phase and finishes with a gradual “washout” of the structure grains. The formation of small caverns was observed [4, 7] after three hours of cavitation. On the surface of F4S15 they are somewhat smaller than on the surface of F4. No formation of “silver cracks” and deep cavities was observed.

The analysis of wear rate kinetics for fluoroplastics (weight loss per unit worn surface area per unit time, $\text{kg}/(\text{m}^2 \text{ h})$) has shown (Figs. 2a, 2b) that for fluoroplastic F4 the maximum wear rate in an acidic medium ranges within 1.3 h, in neutral and alkaline media the maximum corresponds to about two hours of cavit-

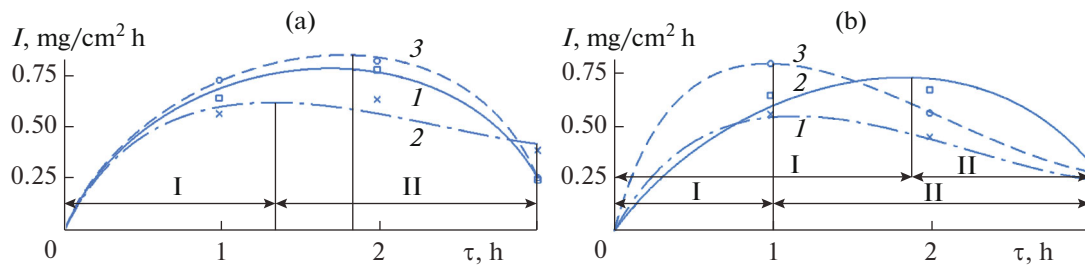


Fig. 2. Wear rate kinetics for fluoroplastics: (a) fluoroplastic F4; (b) impact-resistant fluoroplastic F4S15; (1) neutral medium; (2) acidic medium; (3) alkaline medium.

tion time. For fluoroplastic F4S15, the maximum wear rate corresponds to one hour in alkaline and acidic media, whereas in a neutral medium it is about two hours. At the same time, after three hours of cavitation, the wear rate of fluoroplastic F4 in neutral and alkaline media is the same and amounts to $0.27 \text{ kg}/(\text{m}^2 \text{ h})$, whereas in an acidic medium it is slightly higher amounting to $0.46 \text{ kg}/(\text{m}^2 \text{ h})$. For fluoroplastic F4S15, this parameter is almost the same for all the tested media and amounts to $0.34 \text{ kg}/(\text{m}^2 \text{ h})$ (Fig. 2b). After attaining maximum wear level, the $I-\tau$ curves exhibit a linear decrease in the surface destruction intensity (wear rate), this is especially inherent in fluoroplastics F4 and F4S15 in an acidic medium (Figs. 2a, 2b).

The corrosion resistance of fluoroplastics in most aggressive media can be explained by a high chemical resistance of the C–F bond, which is the strongest among all the carbon bonds with other elements known in organic chemistry. The large size of fluorine atoms and their spiral arrangement around the carbon chain lead to the prevention of chemical attacks to –C–C– bonds in the macromolecule. In addition, the symmetric arrangement of fluorine atoms among each other and their spiral arrangement with respect to the carbon chain determine a minimum strength of intermolecular forces and an insolubility of fluoroplastics in all the solvents [3]. At the same time, under the conditions of cavitation, a local increase in temperature to 1273 K and higher [7], and an increase in pressure to 1000 Pa could occur. Taking all of this into account, as well as a complicatedly strained surface condition [7], chemical reactions between the fluoroplastic and the medium can be initiated.

It is known that the fluoroplastics can dissolve in benzene, toluene, xylene and in some other solvents at high temperatures and pressure. They can also swell in certain media at a temperature higher than 573 K [3]. In addition, the presence of hydrogen atoms H and chlorine atoms Cl with their subsequent entry into the main molecular chain of fluoroplastics results in a decrease in their heat resistance. The presence, for example, of halogen atoms in the polymeric chain at a temperature of 643–663 K results in HF detachment and to the breaking of the polymeric chain, which

leads to a decrease in the physicochemical properties of the polymers. The fact that the formation of chlorine and hydrogen atoms in the course of cavitation in neutral and acidic solutions is quite possible, as well as that the chemical transformations described above occur, can explain the wear resistance of fluoroplastics, in particular, in neutral and acidic media.

The analysis of fluoroplastic F4 wear rate depending on time $I-\tau$ (Fig. 2a) makes it possible to distinguish two fracture stages I and II that differ from each other not only in wear rate, but also in the sign of its first derivative (at stage I, the wear rate increases, whereas at stage II it decreases). For fluoroplastic F4 in an acidic medium, changing wear rate occurs after 1.3 hours, whereas in a neutral medium the wear rate changes after two hours of cavitation. For fluoroplastic F4S15, the duration of stage I in alkaline and acidic media is one hour, whereas in neutral medium it amounts to two hours. Accordingly, the weight loss at stages I and II is distributed at a ratio of approximately 80 and 20% for fluoroplastic F4 in neutral and alkaline media, respectively, whereas for F4S15—in neutral media. In an acidic medium for F4 and fluoroplastic F4S15, this ratio is 40 to 60%, and for fluoroplastic F4S15 in an alkaline medium, this ratio is 50 to 50%.

It is obvious that such an assessment of the destruction stages according to weight loss depending on time in the course of cavitation-erosion wear is tentative, but at the same time, the presence of these two wear stages is beyond controversy. The plotting of $\log \Delta G-\tau$ (dependence in semilogarithmic coordinates (Fig. 3) also indicates the presence of two wear stages. In this case, the inflection point of the straight lines corresponds to the time of attaining the maximum wear rate (Figs. 2a, 2b). The slope of these lines with respect to the abscissa corresponds to the rate of polymeric surface wear at different fracture stages and is measured by the value of $\tan \alpha$. At stage I $\alpha_1 > \alpha_2$ and $\tan \alpha_1 > \tan \alpha_2$, which means that the wear rate (wear intensity) $v_1 > v_2$.

According to [8–10], the destruction of polymers is based on mechanochemical processes, i.e. the combined action of mechanical stresses and chemical reactions, especially in the presence of a corrosive

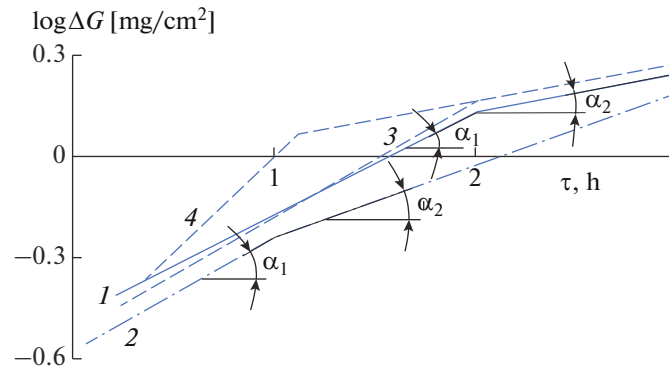


Fig. 3. Weight loss kinetics: (1–3) for fluoroplastic F4 and (4) for impact-resistant fluoroplastic F4S15 (4); (1) neutral medium; (2) acidic medium; (3, 4) alkaline medium.

medium, whereas the thermal destruction caused by the accumulation of local microstructural defects owing to cavitation intensifies the course of chemical transformations [5, 11].

In the case of polypropylene under the cavitation in all the studied model media, the following three fracture stages have been revealed [5]. At stage I, the weakest chemical bonds are broken, as well as the formation of “silver cracks”, whereas their concentration at the sites of intense plastic deformation accelerates the penetration of the working medium deep into the surface of the polymer. This promotes chemical reactions, especially in places where reactive centers in the form of radicals, dangling chemical groups, fibrils, globules, etc. are concentrated. The latter leads to an accelerated destruction of the material surface. Periodic cavitation shockwaves increasingly weaken the binding of these microvolumes between each other and remove them from the surface.

At the second destruction stage, the breakage of strands break and the opening of cracks occurs, their growth, as well as the washing-out of bonded, but already disconnected microvolumes of the polymer are observed. At the same time, the wear rate decreases, since most weight loss occurs at stage I. Thus, at stages I and II, the weight loss amounts up to 80% of the entire weight loss in the course of testing. At stage III, dangling strands are washed out, the surface spherulite is almost completely removed from the surface, and the next spherulite of the polymer takes up the load. The intensity of weight loss approaches a minimum. It should also be noted that the thickness of the spherulite under destruction is equal to the depth of “silver cracks”.

The scheme of the wear rate depending on cavitation time (Fig. 4), plotted in accordance with experimental data from [5], indicates its identity to the scheme for the thermal destruction of polymers in vacuum depending on time, wherein it has been also established that there are three destruction stages that differ in the rate of weight loss [5, 7]. Unlike the ther-

mal destruction in a vacuum, wherein the temperature is a single determining factor, in the course of cavitation exposure to corrosive media, the determining factor consist in a mutual interaction involving temperature T , pressure p , stresses (σ , τ) and chemical reactions between the structural components of the polymer and the medium.

According to the scheme of weight loss rate depending on time (see Fig. 4a), after the evolution of volatile components and low-molecular impurities (segment AB), stage I begins (segment BC). At point C , the first stage of destruction is almost finished. Then the second stage of destruction begins (segment CD) that finishes at point D . Segment DE corresponds to the third stage, i.e. stage III where an almost complete mechanical destruction occurs.

Via the extrapolation of straight lines BC , CD and DE (see Fig. 4a) to the ordinate axis, one can obtain the values of the initial weight of the components ($G^0 = G_I^0 + G_{II}^0 + G_{III}^0$) and the initial values wear rate ($I^0 = I_I^0 + I_{II}^0 + I_{III}^0$).

As already mentioned above, the wear mechanisms at stages I and II are identical and then segments CD and BC can be replaced by common segment DB (Fig. 4a), whereas its extrapolation to the ordinate axis (segment DK) gives the initial value of destruction intensity $I_4^0 = I_I^0 + I_{II}^0$ under the assumption that $I_{III}^0 \approx 0$. The latter follows from our experimental data [5].

In the case of fluoroplastics, owing to their high chemical resistance in the media we studied, the AB segment is absent (Fig. 4b). So, in the range of 473–696 K, the amount of gaseous products that are released is so small that they cannot be registered. At 573 K, the fluoroplastic weight loss amounts to only $2 \times 10^{-4}\%$ per hour, and heating to 663 K also does not lead to any significant weight loss over many hours. Considerable weight changes occur only at a temperature higher than 696 K (0.1% per one hour) [12].

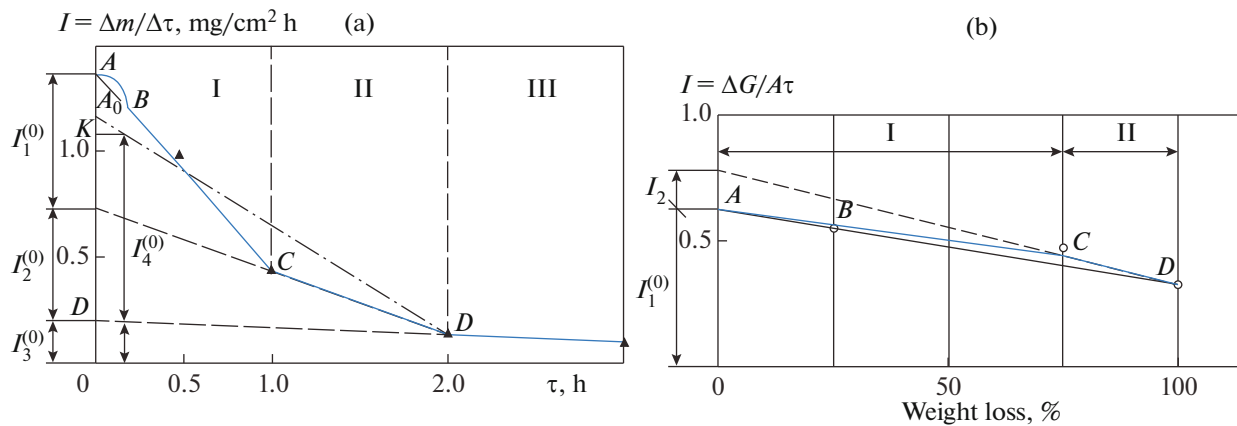


Fig. 4. Scheme of wear rate depending on cavitation time in an acidic medium: (a) polypropylene; (b) fluoroplastic F4S15.

The linear character of the $\log \Delta G - \tau$ and $\log I - \tau$ dependences indicates that it is quite possible to use the kinetic theory of destruction developed by Zhurkov for determining the longevity of fluoroplastics, just as in the case of the cavitation destruction of polyolefins. At the same time, owing to the specific molecular structure, polytetrafluoroethylene (F4) exhibits specific physicochemical properties such as very low friction coefficient, plasticity at a low temperature, etc. The spherulite of PTFE, in comparison with other polymers, is much more defective and friable [3]. Therefore, for fluoroplastics, the determining process consists in breaking intramolecular bonds, rather than in breaking chemical bonds [13].

So, the activation energy required for the breaking of chemical bonds inherent in fluoroplastic F4 is $U_0 = 313$ kJ/mol according to [10], and $U_0 = 345$ kJ/mol according to data obtained by Madorskii and Ellinik. Bartenev indicates the presence of “strong” and “weak” chemical bonds therein. The destruction process is initially determined by the breaking of weak intermolecular internal bonds, which is not accompanied by any significant value of wear rate I at the I-st stage of the surface destruction (Fig. 4b). For impact-resistant fluoroplastic F4S15 in an acidic medium, the difference in wear rate at the I-st stage (straight line AC) and at the II-nd stage (straight line CD) is insignificant, which makes it possible to describe the wear process using single straight line AD (see Fig. 4b).

In the general case, at stage I, the weakest internal intermolecular chemical bonds undergo breaking, as well as the nucleation and accumulation of silver cracks, chemical reactions in spherulites, and microcracks and submicrocracks are formed. It follows from the experimental data that the primary act of polymer destruction consists in the breaking of internal intermolecular bonds, accompanied by appearing free radicals. Further, secondary chemical reactions occur consisting in the conversion of radicals into inactive

products. This period represents a period of accumulation (incubation) of destructed species; it takes, depending on the character of medium, 80% of time in a neutral medium, and 50–75% of time in alkaline and acidic media (Fig. 4b), respectively.

Further (stage II), alongside the increase in the number of broken chains and free radicals that undergo chemical transformations, the products of transformation are washed out by cavitation shockwaves. The destruction intensity thus increases.

Assuming that the decomposition reactions of the structural components of the polymer (by an analogy with thermal decomposition) represent first-order reactions, one can write:

$$G = G_I + G_{II}, \quad (1)$$

where G_I and G_{II} are the weight loss values corresponding to wear rates $I_I = dG_I/dt_I$ and $I_{II} = dG_{II}/dt_{II}$, respectively.

Then

$$I = I_I + I_{II}. \quad (2)$$

For first-order reactions, one can write the following relationship:

$$I_I = K_I G_I, \quad I_{II} = K_{II} G_{II}, \quad (3)$$

where K_I, K_{II} are the reaction rate constants.

According to the theory of chemical reaction kinetics, we have:

$$K_I = C_1 \exp\left(\frac{-E_1}{kT}\right), \quad K_{II} = C_2 \exp\left(\frac{-E_2}{kT}\right), \quad (4)$$

where E_1 and E_2 are the activation energies at stages I and II.

By extrapolating straight lines BC and CD (Fig. 4b) to the ordinate axis, one obtains the values of initial masses and initial fracture intensities:

$$I_1^{(0)} = C_1 G_1^{(0)} \exp\left(\frac{-E_1}{kT}\right), \quad I_2^{(0)} = C_2 G_2^{(0)} \exp\left(\frac{-E_2}{kT}\right) \quad (5)$$

or

$$\begin{aligned}\log I_1^{(0)} &= \log C_1 G_1^{(0)} \exp\left(\frac{-E_1}{kT}\right), \\ \log I_2^{(0)} &= \log C_2 G_2^{(0)} \exp\left(\frac{-E_2}{kT}\right).\end{aligned}\quad (6)$$

In the case of impact-resistant fluoroplastic F4S15 in an acidic medium (Fig. 4b), straight lines *BC* and *CD* can be replaced by one straight line *BD*. This follows from the fact that the destruction mechanism is the same in the two segments and exhibits a mechanochemical character. At stage I, the determining mechanism consists in a chemical destruction, whereas at stage II it consists in a mechanical destruction. For stage II, one can write [13]:

$$I_2 = \frac{K_4}{\tau_{ak}^n}, \quad \log I_2 = -K_4 n \log \tau_{ak}, \quad (7)$$

where K_4 is the coefficient of proportionality; τ_{ak} is the induction period of deformation accumulation (determined experimentally); n is an exponent that is determined by the conditions of external microshock loading in the course of cavitation.

From equations (6) and (7), one can find activation energies E_1 and E_2 depending on the type of medium.

The weight loss in the course of cavitation-erosion wear can be described by relationship $G = e^{a\tau}$ [14] and then the intensity of surface destruction can be expressed as:

$$I = \frac{G}{\tau} = \frac{e^{a\tau}}{\tau}, \quad (8)$$

where $a = \tan \alpha$ is the slope of straight lines $\log G \Delta - \tau$ (see Fig. 3); τ is the test duration time.

Then the general equation for cavitation-erosion wear has the following form:

$$C_1 G_1 \exp\left(\frac{-E_1}{kT}\right) + C_2 G_2 \exp\left(\frac{-E_2}{kT}\right) = \frac{e^{a\tau}}{\tau}. \quad (9)$$

Taking into account equation (7), one obtains:

$$C_1 G_1 \exp\left(\frac{-E_1}{kT}\right) + \frac{K_4}{\tau_{ak}^n} = \frac{e^{a\tau}}{\tau}. \quad (10)$$

Based on the mechanochemical mechanism of destruction of the surfaces of the fluoroplastic at the I and II stages (Fig. 4b), for obtaining tentative data, one can use the equation

$$\frac{K_5}{\tau_{ak}^n} = \frac{e^{a\tau}}{\tau}. \quad (11)$$

By analogy with fluoroplastics, the initial value of the wear rate at stages I and II (Fig. 4a) can be expressed in the following form:

$$\begin{aligned}I_4^{(0)} &= C_4 m_4^{(0)} \exp\left(\frac{-E_4}{kT}\right), \\ \log I_4^{(0)} &= \log C_4 G_4^{(0)} \exp\left(\frac{-E_4}{2.3kT}\right).\end{aligned}\quad (12)$$

Counting upon one mole, the following relationship can be obtained;

$$\log I_4^{(0)} = \log C_4 G_4^{(0)} \exp\left(\frac{-E_4}{2.3RT}\right), \quad (13)$$

where $R = 8.32 \text{ J/(mol K)}$ is the universal gas constant.

At stages I and II, alongside the thermal (thermo-fluctuation) destruction mechanism, the wear process is also determined by a mechanical factor that in this case serves as a catalyst for the wear of the surface spherulite. Therefore:

$$I = I_4 = I_5 + I_{\text{mech}} \quad (14)$$

or

$$I = C_5 G_5 \exp\left(\frac{-E_5}{RT}\right) + \frac{K_4}{\tau_{ak}^n} \quad (15)$$

and then

$$C_4 G_4 \exp\left(\frac{-E_4}{RT}\right) = C_5 G_5 \exp\left(\frac{-E_5}{RT}\right) + K_4 n \log \tau_{ak}^n. \quad (16)$$

The values of activation energies calculated from the foregoing relationships and the experimental data obtained in [5] are as follows: $E_1 = 128 \text{ kJ/mol}$, $E_2 = 231 \text{ kJ/mol}$, $E_3 = 117 \text{ kJ/mol}$, which coincide with the values obtained by the authors of [10]. Somewhat higher values of the activation energy obtained in this work may be explained by the fact that $T = 293 \text{ K}$ has been taken for the calculations, and that local temperature blazing in the course of cavitation have not been taken into account.

Equation (16) also makes it possible to quantify the mechanochemical wear factor in the course of cavitation depending on the conditions of external loading (radiation energy, the amplitude and frequency of vibrator oscillation, a type of medium).

CONCLUSIONS

(1) Experimental data have been obtained for cavitation-erosion wear resistance exhibited by fluoroplastic F4 and its impact-resistant analog F4S15 in acidic, alkaline and neutral media. The kinetics and possible mechanisms of surface wear have been analyzed on this basis for the studied polymers throughout the entire cycle of surface spherulite destruction.

(2) For fluoroplastics, two fracture stages with identical wear mechanisms (mechanochemical) have been revealed, whereas for propylene, three wear

stages have been obtained: wear stages I and II are characterized by a thermochemical destruction mechanism, and stage III is mainly mechanical in the fracture character. Proceeding from this, basic schemes for cavitation-erosion wear inherent in fluoroplastics and polyolefins in corrosive media have been proposed.

(3) Wear rate equations for fluoroplastics and polyolefins are obtained, and initial fracture energies for each wear stage are determined.

NOTATIONS

ΔG	is the weight loss of the material under investigation
I	is the wear rate
MSV	magnetostriction vibrator
F4	polytetrafluoroethylene PTFE, fluoroplastic F4
F4S15	PTFE reinforced with fiberglass, impact-resistant fluoroplastic F4S15

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