An Investigation of the Change in the Properties and Orientation Effects of Phenoplasts of Phenolaniline–Formaldehyde Resol Resin and Kaolin Filler Depending on the Degree of Elastification of the Resin with Nitrile Rubber

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Synopsis

An investigation is made of the dependences of the values of 13 types of standard physical, mechanical, and electric insulation indicators for phenoplasts of phenolaniline–formaldehyde resol resin on the degree of kaolin filling (within the limits of 24 to 47 vol-%) and different degrees of elastification (expressed in SKN-40 nitrile rubber content ranging from 0 to 40 vol-% as to the polymer).   After expressing these dependences as a function of the recalculated average thickness of the adsorption coating of elastified resin on the filler particles, it is established that the characteristic periodical recurrence of concave and convex sectors in the curves for the individual indicators with the non-elastified phenoplasts, which is due to the orientation effects, is preserved also with the elastified phenoplasts.   A method is elaborated of determining the average weighted value of the average thickness of the adsorption coating in the points of the orientation extremums of these curves.   It is found that the places of the individual extremums change relatively little with the increase in the degree of elastification.   A considerable change in the relative values of the orientation effects is observed after the point of phase inversion of the elastified resin (18 to 20 vol-% of SKN-40 as to the polymer) in connection with the sharp increase in the mobility of the resin globules.   The author believes that the proposed mechanism of change of the properties with the filling will be observed, under definite conditions, also when filling with powdered fillers other polymers or blends of polymers, in so far as the latter are made up of supramolecular structural formations with a definite statistical distribution of their average sizes.

INTRODUCTION

Results of thermomechanical and physicochemical investigations of filled polymers show that in the interaction with the surface of the particles of the filler, the polymer's secondary structural formations take part.[1] It is proved that the experimentally established increase in the average times of relaxation of the filled polymers, compared with the nonfilled ones, is due in the first place to a decrease in the mobility of the supramolecular structures.   With the decline in the density of the space lattice of the cross-linked polymers and the increase in the mobility of the larger structural

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units, a higher degree of influence of the surface of the filler and hence a more important change in the average relaxation time and the indicators of the filled as to those of the nonfilled polymers can be expected.

The decrease in the brittleness of the crosslinked phenol–formaldehyde resins and the improvement in their elastic properties is achieved through the plasticizing or tempering[2] or elastification[3] with rubber elastifiers compatible with them.[4] According to experimental data, the solubility of phenol resins in vulcanizates with different rubbers declines the larger the difference in the parameters of solubility of the two polymers and the higher the resin's molecular weight.[5] Phenol resins are compatible with butadiene–acrylonitrile rubbers. Comparing the solubility parameters of phenol resins with those of nitrile rubbers, it can be assumed that the thermodynamic compatibility of these resins with butadiene–nitrile rubber appears at a level of some 36% of nitrile group content, while the remaining mixes are two-phase. After the curing and the solidification of the phenol resin, however, the possibility of obtaining a thermodynamically one-phase system will be restricted owing to the sharp decline in the solubility of the resin in the rubber.[5]

According to Kuleznev et al.,[6] a given polymer dispersed in another polymer but thermodynamically incompatible with it can exist simultaneously and partially in three states: first, in the state of a genuine solution; second, in the state of a quasi-solution, i.e., of a quasi-dissolved part, as a result of the segmental solubility of the transition layer of the phase interface; and third, in the state of a separate phase. On the basis of experimental and literature data, the same authors believe that the share of the genuinely dissolved polymer is insignificant and amounts to between a fractional value and several per cent, while the share of the quasi-dissolved polymer in the phase interface can reach several tens per cent. Taking into consideration the insignificant mutual solubility of the polymers, Gul and Kuleznev[7] presume that their different affinity with one another expresses itself not in a molecular but in a different segmental solubility which leads to the emergence of a transition layer of a different thickness in the phase interface.

According to Schwarz and Dinsburg,[5] in the majority of the cases as a result of the blending of rubbers with thermosetting resins, a two-phase system is created in which, in the basic mass of the rubber or the modified rubber and resin polymer obtained as a result of the chemical interaction of the rubber with the resin, a second phase of resin or rubber and resin highly organized structures is dispersed.

Electron microscope observations[8] of the structure of sulfur vulcanizates of SKN-40 nitrile rubber and phenolformaldehyde Novolak resin blended with the rubber on mills at a temperature of 60° to 80°C have shown the existence of resin particles of the size of several microns, in compact contact with the adjacent rubber and resin space. In the presence of urotropine, the resin particles disperse to smaller sizes.

According to Tumanov et al.,[9] the microstructure of the mill-blended system of phenolaniline–formaldehyde resin No. 214 and SKN-40 nitrile rubber in a ratio of 1:1 varies from a fibrillous to a globular structure depending on the temperature of the mills (45° to 95°C) and the subsidiary thermal treatment. According to the data from the electron microscope investigations carried out by the authors above, the blend obtained at a temperature of the mills of 90° to 95°C and then heat treated at 180°C possesses an oriented fine globular structure with a globule diameter of around 300 Å.

According to Schoon's electron microscope investigations,[10] the microstructure of the butadiene–acrylonitrile rubber Perbunan 3810 (with 38% nitrile groups) consists of macromolecules having a more or less ball-shaped form and sized between 30 and 100 Å (average value 55 Å).

According to the developed conceptions,[1] in the case of filled plasticized polymers, too, an interaction of the secondary supramolecular structures with the surface of the filler takes place, but this interaction changes under the effect of the introduced plasticizer. The latter can exert an influence on the mobility of the chains and take part in the interaction with the surface of the filler (a competition between the polymer and the plasticizer for places on the surface of the filler taking place). According to these conceptions, the process of plastification of the filled polymers is determined by two simultaneously acting effects, the interpacket plastification[11] at the polymer–filler interface resulting in a disturbance in the interphase links, and a plastification determined by the disturbance of the type of polymer–polymer intermolecular links. According to the experimental data, at the beginning the first effect exerts a stronger influence, but with the further increase in the plasticizer content the second effect begins to play a more inportant role.

In an older work,[12] it has been established that the properties of phenoplasts on the basis of phenolaniline–formaldehyde resol resin and kaolin filler change periodically with the filling, depending on the average thickness of the adsorption resin coating on the filler particles, this being due to positive and negative orientation effects. These effects are explained by the periodical change in the physical structure of the adsorption coating, with the increased filler amount, and hence the passing through a minimum and a maximum of the number of oriented resin globules. Admitting by analogy with the calculations in reference[13] that the resin preserves a steady solvatation volume[14] as to the phenoplasts' total volume, the calculated average thickness of the adsorption coating in the points of the orientation extremums is an aliquot of half the average diameter of the resin globules.[12] It can be expected that with the elastification of the resin with nitrile rubber and the increase in its molecular mobility, changes will intervene in the size and the place of the orientation extremums in filling.

The aim of the present study is to investigate with the phenoplasts based on phenolaniline–formaldehyde resin and a kaolin filler the change in

the properties and the orientation effects with the filling depending on the degree of elastification of the resin with SKN-40 nitrile rubber.

EXPERIMENTAL

In addition to the materials described in a previous work,[14] butadiene–acrylonitrile rubber SKN-40 (GOST 7738-65) and a curing combination of sulfur and Altax to the extent of 2.5% as to the rubber were utilized. Besides the phenoplasts of our previous work,[12] phenoplasts were elaborated and tested with a degree of elastification (SKN-40 nitrile rubber content) of up to 40% (volumetric) as to the polymer and a kaolin filler within a ratio of 24 to 47% (volumetric) as to the blend as a whole. A total of 29 combinations, indicated in Figure 1, were investigated.

The elaboration of the blends was done on laboratory mills, under the following sequence of operations: first, the nitrile rubber was fed to the mills and then the powdered blend of the remaining ingredients was added. Temperature, milling time, and the methods of preparation of the samples and of their testing are described in reference.[14] The time of pressing of the rubber containing phenoplasts is 1.5 min per 1 mm sample thickness.[15]

Diagrams and Spatial Figures for the Individual Indicators

The determined values of a given indicator of the blends were plotted onto the respective points for their composition on the Gibbs diagram to
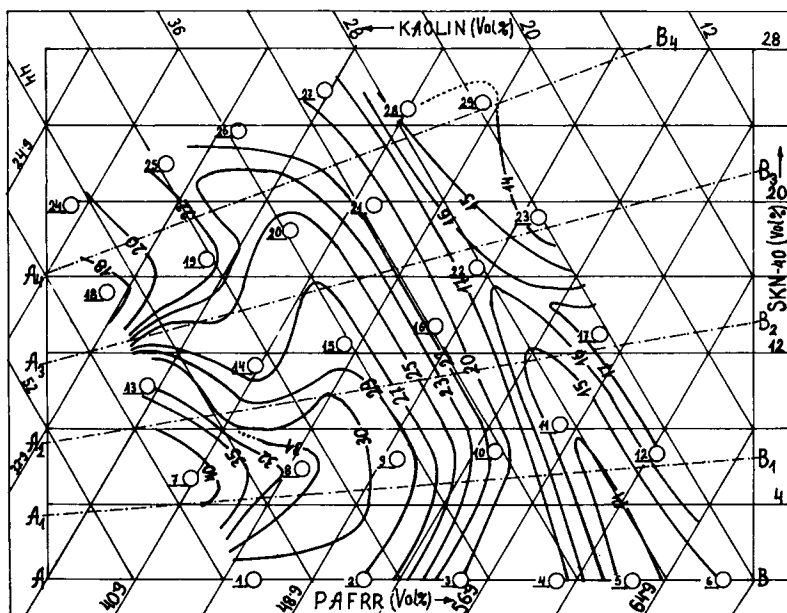

Fig. 1. Gibbs diagram of dielectric constant with indication of the composition of the individual phenoplasts.
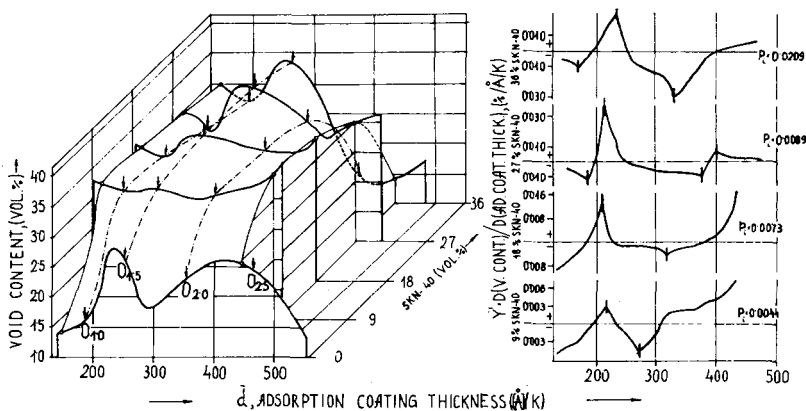
Fig. 2. Dependence of void content on thickness of the adsorption coating and degree of elastification (to the left, conventional curves; to the right, differential curves).
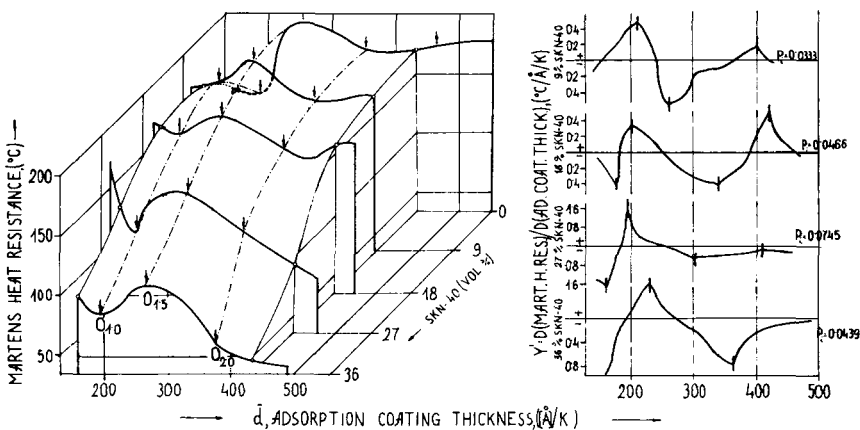

Fig. 3. Dependence of Martens heat resistance on thickness of the adsorption coating and degree of elastification (to the left, conventional curves; to the right, differential curves).

a definite scale with the aid of a device with vertical scale pins, and the resulting spatial figure was cartographed on the base (Fig. 1). From the diagrams thus obtained, for every indicator the dependences of the indicator value were determined on the volumetric concentration of the filler for elastification levels E = 0, 9, 18, 27, and 36 vol-% rubber in the polymer share (the dotted straight lines AB, A_1B_1, A_2B_2, A_3B_3, and A_4B_4 on Fig. 1).

In Figures 2–14 (to the left), the spatial figures are given for the values of the individual indicators for the various degrees of elastification depending on the average thickness $\bar{d}$ of the adsorption coating of the two polymers around the filler particles. This average thickness is given on the abscissa as a value recalculated from the volumetric concentration and certain indicators of the filler in [Å], which value must be divided by the coefficient K specific for the given volumetric concentration of the filler for the degree
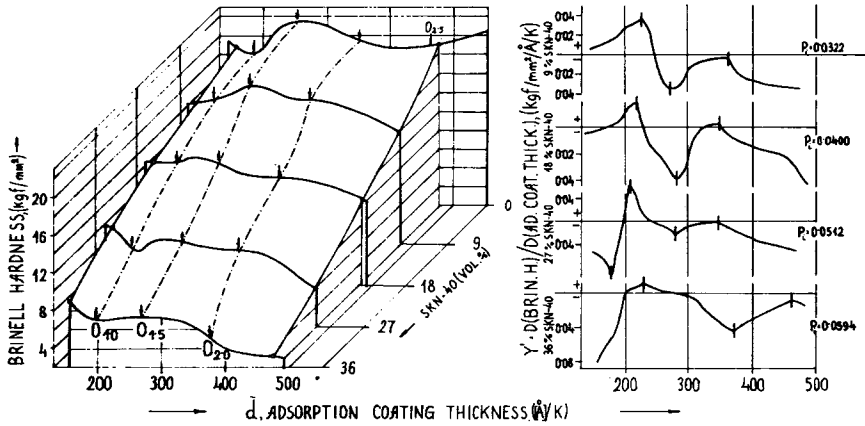
Fig. 4. Dependence of Brinell hardness on thickness of the adsorption coating and degree of elastification (to the left, conventional curves; to the right, differential curves).
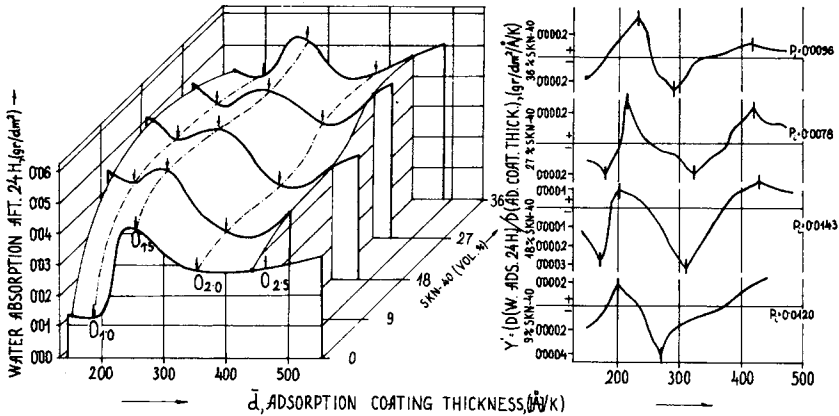

Fig. 5. Dependence of water absorption after 24 hr on thickness of the adsorption coating and degree of elastification (to the left, conventional curves; to the right, differential curves).

of coating of the surface of the filler particles.[14] For the time being, no direct methods for calculating this coefficient are known, that is why in the present work the average thickness of the adsorption coating is recalculated from the volumetric concentration of the filler and is given in $\text{Å}/K$ values. Since in individual cases a certain extrapolation was made, the individual curves of Figures 2–14 (to the left) are limited on both sides with points with equal values for the average adsorption coating thickness.

By the curves for the indicator value for every degree of elastification, differential curves with a mirror differentiator[16] were drawn, which are given in Figures 2–14 (to the right) and on the basis of which the places of the orientation extremums are given, as described in reference 12.
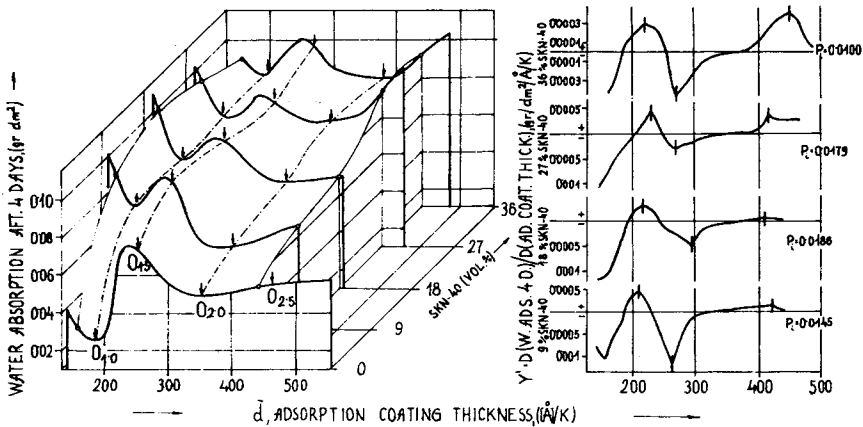
Fig. 6. Dependence of water absorption after four days on thickness of the adsorption coating and degree of elastification (to the left, conventional curves; to the right, differential curves).
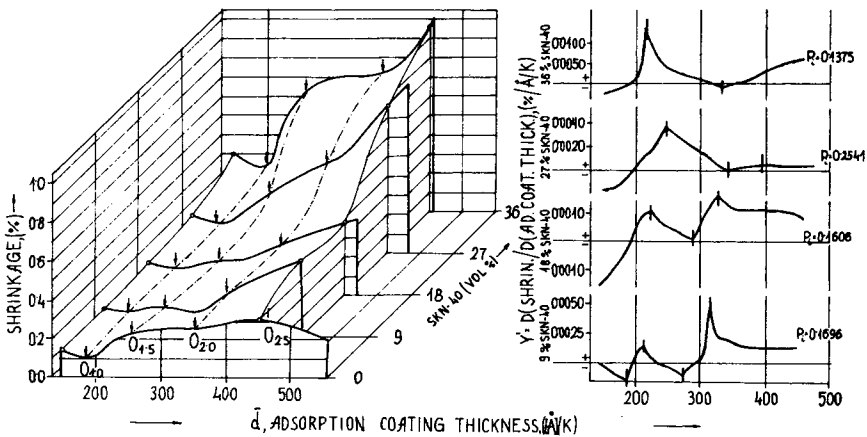

Fig. 7. Dependence of shrinkage on thickness of the adsorption coating and degree of elastification (to the left, conventional curves; to the right, differential curves).

Calculation of the Sensitivity of the Standard Test for a Given Degree of Elastification

For a given degree of elastification E the sensitivity H_i of the standard test to the change in the value of the indicator i within the selected filling range was calculated by[12]

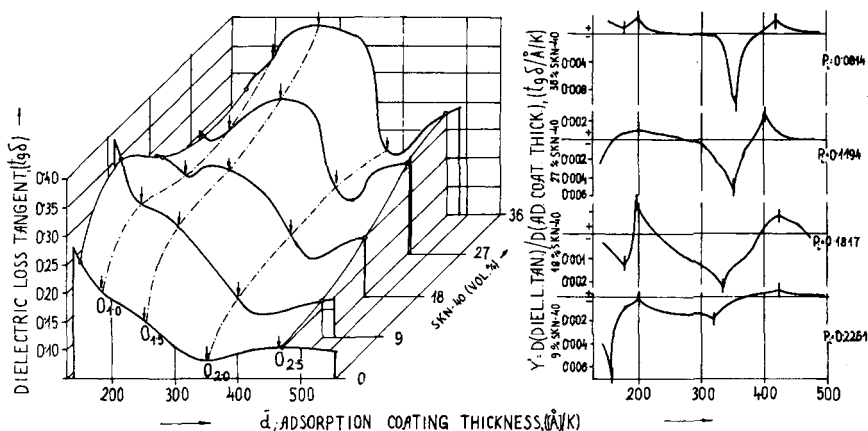$$H_i = \frac{N_{\max} - N_{\min}}{\dfrac{\sum\limits_{m=1}^{n} \mathfrak{I}_{m,\beta}}{n}} \qquad (1)$$

Fig. 8. Dependence of dielectric loss tangent (tg δ) on thickness of the adsorption coating and degree of elastification (to the left, conventional curves; to the right, differential curves).
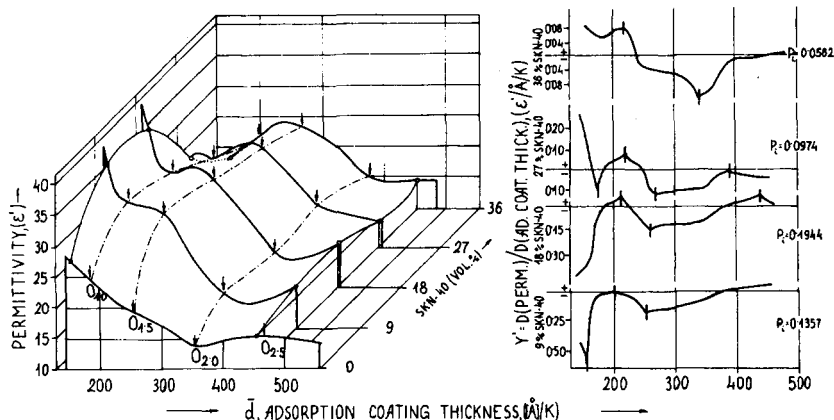

Fig. 9. Dependence of permittivity (ϵ') on thickness of the adsorption coating and degree of elastification (to the left, conventional curves; to the right, differential curves).

where $N_{\max}$ and $N_{\min}$ are the maximum and minimum values of the indicator within the selected filling range for the given degree of elastification E; $\mathfrak{z}_{m,\beta}$ is the confidence interval for the value of the indicator of m-blend for a fiducial probability of $\beta = 90\%$;[17] n is the number of the individual blends with a confidence interval $\mathfrak{z}_{m,\beta}$, which are closest to the straight lines passing through the respective degree of elastification: for $E = 0, n = 6$; for $E = 9\%$, $n = 11$; for $E = 18\%$, $n = 11$; for $E = 27\%$, $n = 10$; and for $E = 36\%$, $n = 10$ (Fig. 1).

Calculation of the Average Weighted Value of the Average Thickness of the Adsorption Coating in the Points of the Orientation Extremums

For a given degree of elastification E, the values of the average thicknesses of the adsorption coatings expressed in $\text{Å}/K$ for the individual orientation
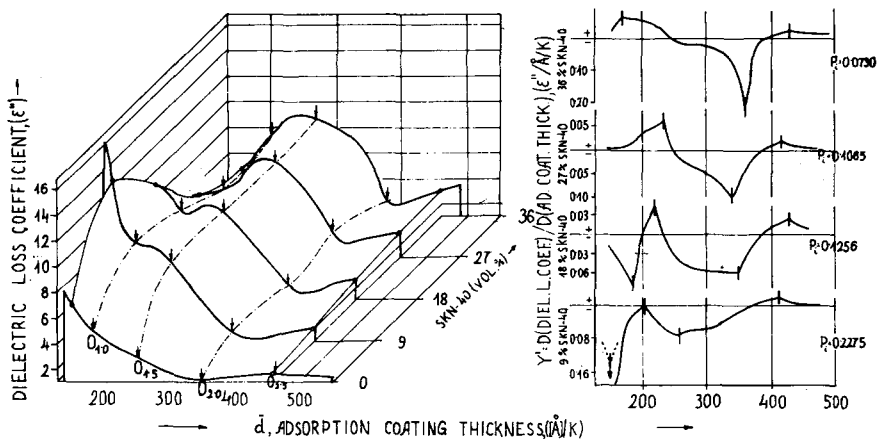
Fig. 10. Dependence of dielectric loss coefficient (ϵ'') on thickness of the adsorption coating and degree of elastification (to the left, conventional curves; to the right, differential curves).
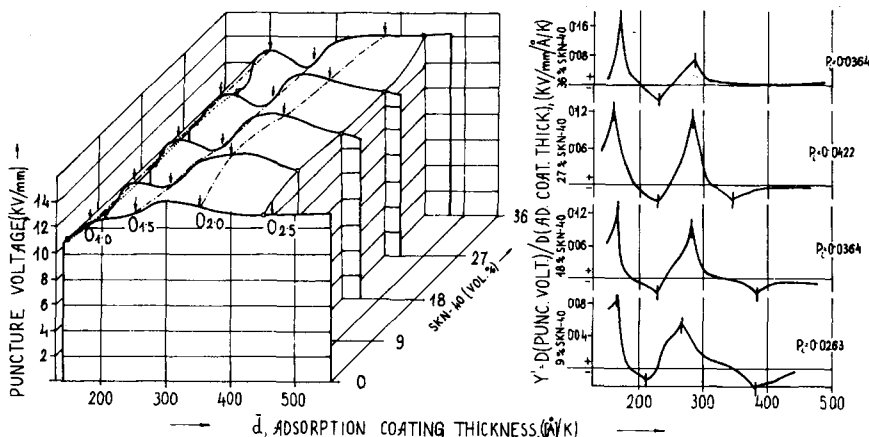

Fig. 11. Dependence of puncture voltage (E_p) on thickness of the adsorption coating and degree of elastification (to the left, conventional curves; to the right, differential curves).

extremums determined by the curves of Figures 2–14 (to the right) show certain differences, because they are determined through standard tests of different sensitivities. Thus, these values are not of an equal worth and should enter in the most likely value with a definite weight P_i. In the absence of systematic errors in their determination, we assume that this weight is connected with the respective sensitivity through the expression

$$P_i = \frac{H_i}{\sum\limits_{i=1}^{q} H_i} \qquad (2)$$

where H_i is the sensitivity of the standard test of the i indicator for a given degree of elastification E, while the denominator of the above expression
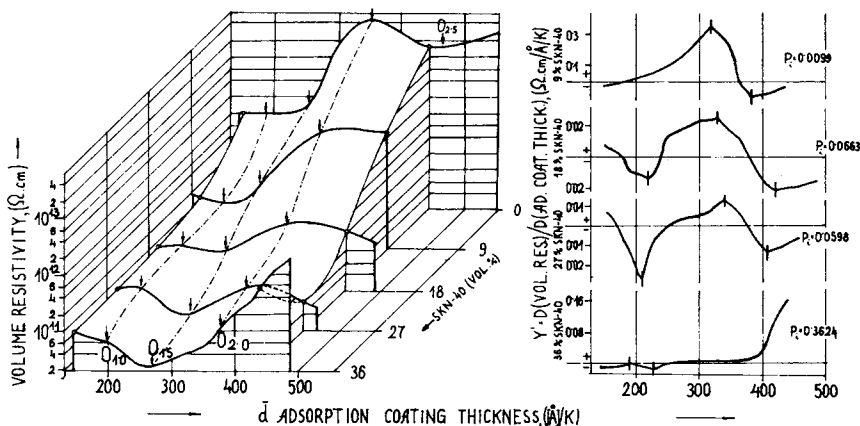
Fig. 12. Dependence of volume resistivity (ρ_v) on thickness of the adsorption coating and degree of elastification (to the left, conventional curves; to the right, differential curves).
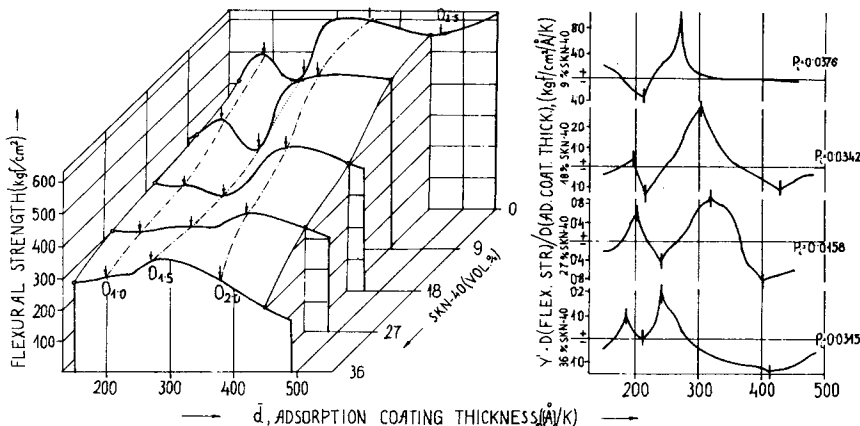

Fig. 13. Dependence of flexural strength on thickness of the adsorption coating and degree of elastification (to the left, conventional curves; to the right, differential curves).

gives us the sum total of the sensitivities of all the q indicators determined by us for the same degree of elastification E.

The weights P_i, calculated in this way for every individual degree of elastification E for the curves of Figures 2–14 are plotted to the right of the corresponding differential curves.

The most likely one, the average weighted value $\bar{d}_w$ (w = weighted) of the average thickness of the adsorption coating for a definite orientation extremum O_p and a given degree of elastification E will be given by the expression

$$\bar{d}_w = \frac{\sum\limits_{i=1}^{q} \bar{d}_i P_i}{\sum\limits_{i=1}^{q} P_i} \tag{3}$$
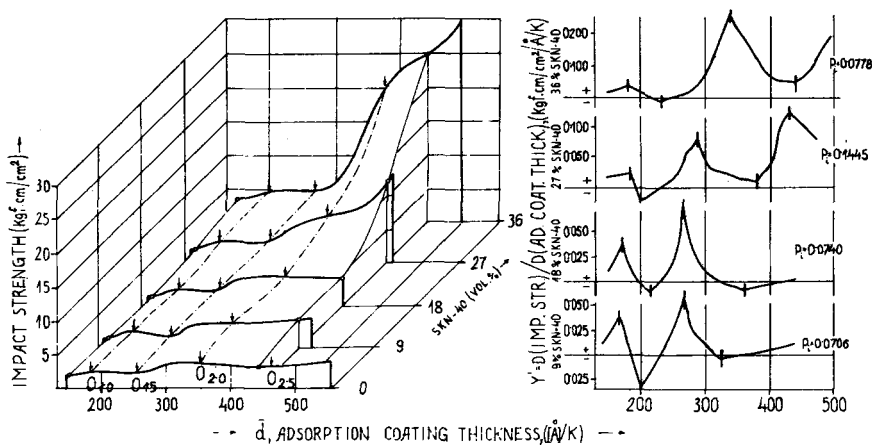
Fig. 14. Dependence of impact strength on thickness of the adsorption coating and degree of elastification (to the left, conventional curves; to the right, differential curves)

where $\bar{d}_i$ is the average thickness of the adsorption coating in the point of the extremum O_p for a degree of elastification E determined by its i indicator; and P_i is the weight of the i indicator for the same degree of elastification E.

If we substitute in formula (3) the values calculated by us of the investigated indicators in $\text{Å}/K$ for $\bar{d}_i$, and if we assume that the coefficient K is preserved constant within the range of these values, we obtain both the average weighted value of the average thickness of the adsorption coating for the extremum O_p and the degree of elastification E in $\text{Å}/K$.

RESULTS AND DISCUSSION

The calculated average weighted values of the average thicknesses of the adsorption coatings in the points of the orientation extremums for the various degrees of elastification in $\text{Å}/K$ with the corresponding 95% confidence intervals (the dotted line) are given on Figure 15 (to the left).

Their place is indicated with the aid of perpendicular arrows also on the spatial Figures 2–14 (to the left), where the points of one and the same orientation extremum are connected by a common dotted line.

As can be seen from the spatial figures, the characteristic periodical recurrence of concave and convex sectors of the curves depending on the filler content with the nonelastified phenoplasts[12] repeats itself also with the elastified phenoplasts.   The orientation effects, which depend on the mobility of the globules, vary with size, but this change is not proportionate to the degree of elastification.   In Figure 15 (to the right), the values of the elasticity modulus of phenoplasts with 32 vol-% kaolin content are given for degrees of elastification of up to 40 vol-% nitrile rubber content, and the values of certain indicators in the points of the more important orientation extremums for degrees of elastification of up to 36 vol-% nitrile rubber content are also quoted.   The saltatory change in the properties of the
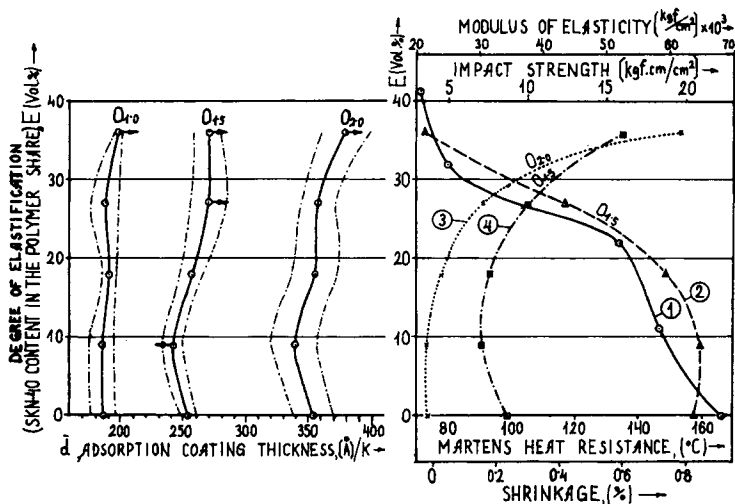
Fig. 15. Dependence on degree of elastification of: left, $\bar{d}_w$ in the points of the orientation extremums; right, (1) modulus of elasticity, (2) Martens heat resistance, (3) impact strength, (4) shrinkage of the phenoplasts.

elastified phenoplasts at a degree of elastification of about 18 to 20 vol-% nitrile rubber content tells of an inversion of the phase structure of the blend of the two polymers.[5,7] Up to this rubber content, the mechanical indicators change less, and it can be assumed that the resin is the dispersion medium. After the inversion, the rubber is the dispersion medium (or the rubber–resin substance, for that matter), which is proved by the sharp decline in the elasticity modulus and the Martens heat resistance, as well as by the sharp increase in the impact strength and the shrinkage (Fig. 15, to the right).

When elastifying phenoplasts on the basis of phenolaniline–formaldehyde resol resin and a kaolin filler, the orientation extremums change their place, but this change, expressed in volumetric concentration of the filler, is relatively small. The data show that at a degree of elastification of 36 vol-% nitrile rubber content, the orientation extremums compared with those of the nonelastified phenoplasts are shifted by a 1.2 to 1.3 vol-% lower filler concentration.

In the field of a given orientation extremum (the individual curves of Fig. 15, to the left), it can be considered that the coefficient K for the degree of coating of the filler surface is kept constant owing to the proximity of the binding agent and the small difference in the volumetric concentration of the filler. Under this assumption, the average weighted values of the average thicknesses of the adsorption coatings for a degree of elastification of 9, 18, 27, and 36 vol-% nitrile rubber content were successively compared through the null hypothesis[18] with the corresponding value for the nonelastified phenoplasts. With the calculated standard deviations for

these values, the critical magnitude of the criterion is exceeded and the null hypothesis is rejected for the points indicated by a horizontal arrow on Figure 15 (to the left); the differences of the values of the remaining points in respect of the corresponding value of the nonelastified phenoplasts can be considered as nonessential.

On the basis of the precision of the conducted investigations and their analysis, it can be considered that up to the inversion of the phases, the thicknesses of the adsorption coatings in the points of the orientation extremums remain constant; or, if judged by the best expressed orientation extremum $O_{1.5}$, they show a tendency to decline up to a degree of elastification of 9 vol-% nitrile rubber content, after which they increase (Fig. 15, to the left). According to the calculations, the average diameter of the globules, which is 240 Å for the nonelastified phenoplasts,[14] increases for a degree of elastification of 36 vol-% nitrile rubber content, as follows: for an extremum of $O_{1.0}$, to 254.47 Å; for an extremum of $O_{1.5}$, to 255.25 Å; and for an extremum of $O_{2.0}$, to 257.37 Å.

In the light of the existing conceptions,[1,5,7,11] two processes develop simultaneously when obtaining nitrile rubber elastified phenoplasts: first, competition for places on the surface of the filler between the globules of the resin and the rubber and creation of a growing area of adsorption coating from the macromolecules of the rubber; and second, owing to the low real solubility of both polymers in one another, a process of interglobular plastification of the resin takes place, i.e., a penetration of the rubber into the resin's interglobular spaces developes and new globules with a surface rubber[8] or transition layer[7] are formed. The first process, which is predominant at the beginning, leads to a decrease in the average thickness of the adsorption layer because of the smaller size of the rubber macromolecules.[10] After this, the second process will become predominant, which involves the increase in the average thickness of the adsorption layer because of the increase in the average diameter of the globules.

In conclusion, the author believes that the character of the curves for the change in the properties of the polymers when filled with powdered fillers by the mechanism suggested in our former work[12] will be observed also in other polymers or polymer blends in so far as they are made up of supramolecular structures with a definite distribution of their average diameters. This character will be determined mainly by the summary action of the two factors as follows: the chemical factor, reflected in a smooth curve, which will depend on the difference in the chemical structure of the ingredients and their volumetric concentration; and the physical factor, reflected in a periodical curve, which will depend on the physical structure of the filled polymer (or polymer blend), the thickness of the adsorption coating of supramolecular structures, and the orientation effects.

In calculating the average thickness of the adsorption layer, we proceeded from the constancy of the volume of the polymeric solvatation coatings in respect of the total volume of the filled polymer, considering the particles of the filler with adsorption coating as particles of a new kind of filler.[14]

According to our considerations, the particles of the new kind of filler have the same form as the ones of the initial filler, but are larger in size, because it is believed that the rheological and mechanical properties of the thin surface layer of resin come close to the properties of the solid filler.[13] In the more general case, however, the constancy of the volume of the solvatation coatings will be preserved under certain conditions owing to the following considerations:

According to Lipatov and Sergeeva,[19] the strong intermolecular interaction between the polymer molecules leads to a spreading of the effect of influence of the filler's surface in the volume of the polymer. The authors deem it expedient to differentiate between the concept of the polymer's adsorption layer, with which a portion of the segments of the polymer chains are in interaction with the surface, and the polymer's boundary layer, the properties of which are changed under the influence of the surface of the filler compared with the properties of the polymer in the volume.

Sagalaev and Simonov-Emelyanov[20] dwell on the interphase layer in filled polymers. This layer, which by definition is close to the boundary layer, in extreme cases either does not form at all, or contains the entire polymer component. According to these authors, its thickness has no clear boundaries and is an average value determined mainly by the thickness of the polymer's adsorption layer measurable by various experimental methods.

According to Rehbinder,[21,22] the physical structure of polymers filled with dispersion fillers depends on the concentration of the filler, the lyophililyty and the degree of surface adsorption activation of the filler particles, and the latter's form. At higher levels of volumetric filling of the polymer and a sufficient lyophilyty of the surface of the filler particles to the given polymer, the latter passes into a state of a strongly consolidated structurized adsorption-solvate film on the surface of the particles. For lower degrees of filling and a mosaic structure (i.e., an incomplete adsorption surface activation) of the particles of the filler, and owing to their anisodiametricity in particular, is characteristic the formation of chain-and-network coagulation structures. They are formed through the adherence of the filler particles through thin interlayers of polymers between them and do not constitute compact aggregates, but rather fine chains and nonordered space lattices, loose carcasses of primary particles, and their chains or aggregates. These structures are the looser the lower the degree of filling.

In connection with the above, it can be expected that even with a complete surface adsorption activation of the filler particles, the solvatation coatings will preserve a constant volume up to a thickness of the adsorption coating within the limits of the boundary interphase layer. With further reduction in the concentration of the filler, the volume of the solvatation coatings compared to the total volume of the filled polymer will increase in view of the possible twirling or shifting of the new kind of particles owing to the reduced viscosity of their surface. Again, in the presence of conditions leading to the creation of coagulation structures, the volume

of the solvattaion coatings compared to the total volume of the filled poly-
mershould increase in proportion to the looseness of these structures.

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