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Determination of Compatibility of Polymer Systems and Study of Chemical Properties of their Mixtures

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Abstract

We have obtained mechanical strength of materials as a result not by synthesizing new polymers, but by using industrial polymers. We have developed a technology for obtaining new composite materials based on industrial multi-tonnage polymers of high-strength polyethylene (HSPE), polyvinyl chloride (PVC), polyurethane thermoplastic elastomer (DUTEP), butyl rubber (BR), chlorocarboxylate polyethylene (CCPE), ethylene propylene rubber (EPR). To obtain a composition from these polymers, their mixture compatibility was first determined using the most modern methods. Thermodynamic compatibility of the butyl rubber-polyethylene (BR-LDPE), polyvinyl chloride, HSPE, polyurethane thermoplastic elastomer, PVC-chlorocarboxylate polyethylene (CPE), (CREP-VC) systems was studied using inverse gas chromatography methods, and structural features of these systems were revealed using IR spectroscopy and polarization microscopy. The study demonstrated that the incorporation of polyfunctional groups such as chlorine, hydroxyl, and carboxyl markedly influences the compatibility mechanism and provides a consistent set of enhanced properties, including improved adhesive performance, greater tensile and tear strength, and increased resistance to thermal and chemical degradation. These improvements arise from intermolecular polar interactions between macromolecular chains. The solubility parameters of four polymers with varying degrees of polarity were evaluated using turbidimetric titration (TDT) and inverse gas chromatography techniques. Based on the temperature dependence of the solubility parameter derived from sorbent retention times, the parameter value normalized to 25 °C was calculated and compared with that obtained through the TDG method at the same temperature. The results indicated that the discrepancy between the solubility parameters determined by the two methods increased with higher polymer polarity. This phenomenon is attributed to the fundamentally different states of the examined polymer–solvent systems.

Keywords: Polymer, macromolecules, compatibility, high-strength polyethylene (HSPE), polyvinyl chloride (PVC), polyurethane thermoplastic elastomer (PUTE), butyl rubber (BR), chlorocarboxylate polyethylene (CCPE), ethylene propylene rubber (EPR). solubility parameter, sorbent, stability, polyfunctional groups

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INTRODUCTION

Currently, at the industrial level, the challenge of producing polymeric materials with properties suitable for modern applications is addressed through two main approaches: the synthesis of novel polymers and the development of polymer blends or composites, in which the characteristics of individual components combine in an additive manner to enhance overall performance [1-3]. In this regard, we have developed various polymeric compositions in the existing technological conditions and technological equipment, significantly simplifying the technological cycle of processing, radically reducing the weight and dimensions of products and, of course, their cost. is relevant [4-7]. Today, an important task for researchers is the main problems of processing polymer composite systems with polyfunctional properties based on polyolefin and heterochain polymers, providing high physical and mechanical properties of products operating in extreme conditions. Therefore, we believe that the task of the study is to create polymer systems with the required set of operationally important properties. To achieve this, it is necessary to determine the compatibility and interaction parameters of polymer mixtures. In this way, it is possible to determine the polymer systems in which ratios they can be combined with each other, and after determining this, it is possible to say with confidence that these polymer systems in what ratios can be combined [8-12].

The relevance and importance of this problem are determined by the fact that the creation of the theory of multicomponent systems is extremely poorly reflected in the literature, although the importance of using such materials in modern technology can hardly be overestimated [13–16, 5].

The investigation focuses on evaluating the properties of newly developed polymer compositions and examining their processing conditions using existing industrial equipment. It also aims to determine the thermodynamic compatibility patterns of commercially produced polyolefin and heterochain polymers, as well as to explore the feasibility of producing composite materials with substantially enhanced performance characteristics. The integration of various polyfunctional components with optimized properties highlights the practical significance of this research direction [17–19, 8].

Several researchers have proposed chemical and physical modification techniques for individual polymers and their blends to improve the performance of composite materials, achieving notable enhancements in heat resistance under air and vacuum conditions, as well as improved anticorrosive behavior [20–24]. In this context, oil–polymer resin–SPP has been introduced into composite systems intended for manufacturing products that operate under severe mechanical wear, elevated temperatures, high pressures, and significant stresses and deformations [25–29].

Thus, the development of diverse polymer compositions that simplify production and processing technologies while reducing material costs and maintaining required property sets remains highly relevant under current technological conditions [30–34]. This need arises from the still limited theoretical understanding of multicomponent polymer systems and the absence of well-established scientific foundations governing these processes [42-46]. To determine the influence on the mechanism of compatibility of polymers and the possibility of imparting increased adhesive, strength, heat-resistant and corrosion-resistant properties to mixtures, it is necessary to obtain a composition with high properties on their basis; their mixtures must be modified with various polyfunctional groups such as chlorine, hydroxyl, cyanide, carboxyl, ester and others [47-50].

Method

To study the thermodynamic compatibility of binary polymer mixtures, gas chromatography-mass spectrometry (GC-MS) (Figure 1) The advantage of this method is that it combines the capabilities of gas chromatography and mass spectrometry to identify and quantify individual components in complex mixtures [51-69].

A polymer mixture that contained polar carboxyl groups showed that these systems are only partially thermodynamically compatible, despite the presence of both critical compatibility temperatures.

A study of the processes: thermal-oxidative stability of chlorine-containing polymers showed that it depends primarily on the concentration of chlorine-containing groups. The process of thermal destruction occurs with the rupture of C-C bonds in an already hydro chlorinated polymer

To determine the spontaneous nature of the dissolution processes of polymer mixtures and their solubility at constant temperature and pressure, the spontaneous dissolution of polymers was determined by the following formula, [10].

$m\Delta F_{cm,}\Delta H_{cm}, \Delta S_{CM} -$. where, are respectively the changes in free energy, neatly and entropy of mixing the polymer with the solvent.

To determine the critical temperature, which is called the temperature at which the polymer dissolves, was determined from the condition:

$$\Delta F_{cm,}\Delta H_{cm}, T\Delta S_{CM} = 0.$$

From here

$$T_{kp} = \frac{\Delta H}{\Delta S},$$

$T_{kp} - .$ c critical temperature

In the work to study the thermal effect of dissolution processes, which are accompanied by a thermal effect, the total thermal effect during polymer dissolution was determined as follows:

$Qp=k(-Ei_{n}+E_{eb}+2E_{in\text{-}eb.}$

where k is the proportionality coefficient; Ep-r is the interaction energy between polymer molecules; is the interaction energy between solvent molecules; is the interaction energy between polymer and solvent molecules.

To determine the polymer mixtures, the Flory-Huggins model of polymer solution and the phase equilibria of the polymer-solvent system, the following formulas are used

Figure 1. Chromatography-mass spectrometry.

$\Delta F = -T\Delta S = RT\ (n_1 1n\varphi_1 + n_2 1n\varphi_2)$,

where φ_1 и φ_2 – volume fractions of solvent and polymer, respectively; n_1 и n_2 – number of moles of components.

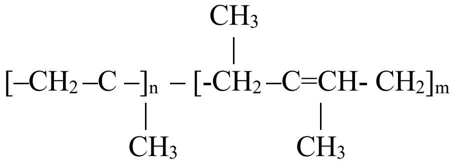
For a two-component polymer-solvent pair, the phase relation was determined using the Gibbs phase rule:

$F = k + 1 - f$, where F– degrees of freedom; k – number of components; f – . number of phases in the system

EXPERIMENT AND DISCUSSION

Calculation of Compatibility for the System of Butyl Rubber-Poly Ethylene of High Pressure (BK-LDPE) At A Temperature of 25°c

To calculate the solubility parameters of each component of the mixture, tabular values of the molar attraction constants of various chemical low-molecular groups were used. The density of the polymers used in the work was determined and the following results were obtained: BR at 25°C ρ = 0.92×103 kg/m3 and the density of LDPE ρ = 0.96×103 kg/m3. The distribution of end groups and molecular weight distributions of systems were not taken into account in the calculation.



$$[-CH_2-\underset{CH_3}{\overset{CH_3}{C}}-]_n-[-CH_2-\underset{CH_3}{C}=CH-CH_2]_m$$

Molar constants of attraction of isoprene

∑F =133,2 + 93 +214 + 28 = 415

M= 5x 12 + 8 x 1 =68

Molar constants of attraction of isobutylene

∑F - CH₂–CH₃ \ /
C
/ \

∑F =133 + 2 × 214 – 93 = 468

M=12 × 4 + 5 × 1 =53

Then the solubility parameter of butyl rubber

$$\delta_{\text{бк}} = 0{,}92/0{,}95\frac{468}{53} + 0{,}05\frac{415}{68} - 8{,}0$$

Solubility parameter HDPE

∑F = 133

M= 1x 12 + 2 x 1 =14

The interaction parameter between the polymers was then calculated using the Hildebrand equation; P is the gas constant, equal to 1.987 col mol-1 deg-1; T is the temperature in °C, and Vp is the reference volume (cm^3/moll), usually taken to be 100 cm^3/mol. For this value of Vp and a temperature of 250C, the Hildebrand equation takes the form:

$$\delta_{\text{бк}} = 0{,}96\frac{133}{14} = 9{,}12$$

In this case = ($\delta_A - \delta_B$)

$$X_{BR}\text{- HDPE} = \frac{(9{,}12-8{,}0)^2}{6} = 0{,}209$$

To calculate the critical temperature value of the, X_{br}-X^{hdpe} the following equation is used:

$$X_{cr} = 1/2[1/X^{1/2}{}_{A} + 1/X_{B}{}^{1/2]}$$

This includes each polymer's degree of polymerization, expressed in terms of comparative volume. The degree of polymerization may be determined from the actual polymerization index (X), provided that the molar volume of the polymer's repeating unit is known, using the following equation.

$$\bar{X}_A = \left(\frac{V}{V_p}\right)_x$$

A fairly good approximation is obtained from the relation

$$X_A = \left(\frac{M_A}{100}\right)_x$$

Where MA is the molecular weight of the polymer.

Thus we have:

$$X_{BR} = \frac{390000}{100} = 3900$$

$$X_{\text{HDPE}} = \frac{320000}{100} = 3200$$

From here, according to the equation for calculating the critical value:

$$(X_{\text{br-hdpe}}) = 1/2 \ x 1/3900^{1/2} + 1/3200^{1/2} = 7{,}18 \text{ x } 10^{-4}$$

Considering that the probability of phase separation of systems is possible, it was necessary to calculate BR-HDPE for different ratios of phase compositions. The results are shown in Table 1.

As can be seen from Table 1, with a decrease in the concentration of any component in the mixture, the spindle solubility parameter increases, while HDPE dissolves in BR significantly better than BR in high-strength polyethylene. The concentrations at which absolute concentration of the original polymers is possible were determined by the variation method. As a result, for HDPE this is 0.08% by weight, and for BR - 0.06%.

The calculation performed proved that the BR-HDPE system is practically incompatible. Therefore, in order to determine the system's critical point on the phase diagram, it is essential to evaluate the Gibbs free energy of mixing (G_{sm}) for the constituent components. The obtained data shows the need to improve these polymers, it is necessary to use functional groups to improve the compatibility of polymer systems.

The calculation was carried out for T = 25ºC, the results of which are presented in Table 2.

Table 1. Solubility parameters of the BR-HDPE system.

F_{br}	$(X_{\text{br-hdpe}})_{sp}$	Fbr	$(X_{\text{br-hdpe}})_{sp}$
0,00	-	0,56	5,3 ×10
0,06	2,03 ×10	0,61	5,51 × 10
0,09	3,61 × 10	0,63	5,99 ×10
0,14	3,15 × 10	0,67	6,50 × 10
0,19	6,23 × 10	0,74	7,67 × 10
0,22	7,14 × 10	0,78	1,51 × 10
0,28	7,24 × 10	0,84	1,78 × 10
0,33	6,61 × 10	0,86	1,9 × 10
0,39	6,36 × 10	0,94	3,18 × 10
0,46	6,24 × 10	1,00	–
0,51	6,21 × 10	–	–

Table 2. The heat of mixing BR-HDPE.

Φ_{br}	G_{mixing}	$\Phi_{нк}$	G_{mixing}
0,00	-	0,59	3,72 × 10
0,05	5,58 × 10	0,63	3,51 × 10
0,10	1,35 × 10	0,66	3,76 ×10
0,15	1,87 ×10	0,72	2,71 × 10
0,20	2,41 × 10	0,77	2,78 × 10
0,25	2,71 × 10	0,82	2,55 × 10
0,30	2,88 × 10	0,84	1,69 × 10
0,35	3,38 × 10	0,89	1,30 × 10
0,40	3,41 × 10	0,98	5,98 × 10
0,45	3,47 × 10	1,00	–
0,50	3,49 × 10	–	–

From the obtained data it is evident that for the BR-HDPE system there is a so-called symmetric case with a lower critical temperature of dissolution and from the principle of additivity we have the system has a critical temperature of dissolution Tcr 230°C.

The obtained data shows that it is possible to conclude:

1. The BR-HDPE mixture is practically incompatible;
2. To obtain partial compatibility

It is possible with a concentration of BR in HDPE up to 0.06% and a concentration of LDPE up to 0.08%.

In the process, the compatibility of the polarity of the components plays a huge role, based on this, we calculated the compatibility parameters for the system of polar components PVC and PUTPE (comparison with the non-polar BR-HDPE system).

Calculation of Compatibility of Thermoelastoplasticified System Polyvinyl Chloride-Polyurethane (Pvc-Putep) At 250c

To calculate the solubility parameters of each component of the mixture, tabular values of the molar attraction constants of various MHGs were used, taking into account the density of PVC at 250C

(ρ25=1.41×103 kg/m_3). In the calculation, the distribution of end groups was taken into account simultaneously and without taking into account the molecular weight distribution.

Single link PVX - $[-CH_2 - CHCl -]$ n

Molar constants of attraction

$-CH_2-$ $-CH = Cl -$

$\sum F = 133 + 28 + 270 = 431$

M=12,2+ 1,3 + 35,5 =62,5

Then the PVX solubility parameter will be:

$\Delta_{pvx} = 1,41$ x431/62,5 =9,72

The solubility parameter of PUTEP, taking into account the density of PUTEP at 25°C (ρ25=1.1×103 kg/m_3).

Single link PUTEP

$[-(CH_2)_6 - OCOHN - (CH_2)_4 -]_n$
$-(CH_2)_6 -$ $- COO-$ $-NH -$ $-(CH_2)_4$
$\sum F = 133{,}6 + 310 + 180 + 133{,}4 = 757$
$M = 12{,}11 + 1{,}21 + 16{,}2 + 14 = 199$
$\Delta_{putep} = 1{,}10 + 1820 + 199 = 10{,}06$

The calculation of the interaction parameter between polymers in the system is performed according to the equation:

$$X_{AB} = V_n / RT\ (\delta_A - \delta_B)^2$$

Where R is the gas constant equal to 1.987 kcal mol-1 dig -1; T is the temperature in K; Vn is the reference volume (cm3/mol), usually taken to be 100 cm3/mol. Thus, for T= T=25^{0}C the equation takes the form:

$$X_{AB} = (\delta_A - \delta_B)^2 / G$$

In this case, for the PVX-PUTEP system we have

$$\Delta_{pvc\text{-}putep} = (10{,}06 - 9{,}72)^2 / 6 = 0{,}019$$

To calculate the critical value (Xpvc-pulp) cr, the equation is used

$$X_{cr} = 1/2[1/X^{1/2}{}_A + 1/X_B{}^{1/2}]^2$$

which includes the degree of polymerization of each polymer expressed in terms of the comparative volume Vn The degree of polymerization can be calculated from the actual degree of polymerization X if the molar volume of the repeating unit of the polymer is known according to the equation

$$X_A = (V/\ V_{p)\ x}$$

A fairly good approximation is obtained from the relation

$$X_A = M_A/100$$

Where MA is the molecular weight of the polymer.

Thus we have:

$$X_{pvx} = \frac{90000}{100} = 900$$

$$X_{putep} = \frac{18000}{100} = 180$$

Отсюда по уравнению для расчета критического значения:
$(X_{pvc\text{-}putep})$ critical temperature $= 1/2 \times 1/X_A (\Phi_A / G_п) + 1/X_в (\Phi B)_{sp}$

The results obtained are shown in Table 3 and 4.

Since 0.019 is much larger than 0.006, i.e. have-put> (have-pulp) cry, then this system should be incompatible over a wide range of compositions.

Using the equation
(XAB) sp= 1/2[1/XA (ФA)sp + 1/XB(ФB)sp]Let us determine the probability of the origin of phase separation, for which we calculate (Xpvc-pulp)sp for different phase compositions. The results of the study are summarized in Figure 2.

Table 3. Values of the spin odal interaction parameter for various phase compositions.

№№ п/п	**PVC**	**($X_{pvc-putep}$) spinodal**
1.	0,00	-
2.	0,05	$2,78 \times 10^{-3}$
3.	0,10	$1,40 \times 10^{-2}$
4.	0,15	$8,64 \times 10^{-3}$
5.	0,20	$6,97 \times 10^{-3}$
6.	0,25	$6,25 \times 10^{-3}$
7.	0,30	$5,93 \times 10^{-3}$
8.	0,35	$5,82 \times 10^{-3}$
9.	0,40	$5,86 \times 10^{-3}$
10.	0,45	$6,02 \times 10^{-3}$
11.	0,50	$6,29 \times 10^{-3}$
12.	0,55	$6,67 \times 10^{-2}$
13.	0,60	$7,18 \times 10^{-3}$
14.	0,65	$7,87 \times 10^{-3}$
15.	0,70	$8,79 \times 10^{-3}$
16.	0,75	$1,01 \times 10^{-2}$
17.	0,80	$1,19 \times 10^{-2}$
18.	0,85	$1,46 \times 10^{-2}$
19.	0,90	$1,92 \times 10^{-2}$
20.	0,95	$2,84 \times 10^{-2}$
21.	1,00	-

Table 4. Values of the spin odal interaction parameter for various phase compositions.

№№ п/п	**PVC**	**($X_{pvc-putep}$)$_{sp}$**
1.	0,00	–
2.	0,05	$2,78 \times 10^{-3}$
3.	0,10	$1,40 \times 10^{-2}$
4.	0,15	$8,64 \times 10^{-3}$
5.	0,20	$6,97 \times 10^{-3}$
6.	0,25	$6,25 \times 10^{-3}$
7.	0,30	$5,93 \times 10^{-3}$
8.	0,35	$5,82 \times 10^{-3}$
9.	0,40	$5,86 \times 10^{-3}$
10.	0,45	$6,02 \times 10^{-3}$
11.	0,50	$6,29 \times 10^{-3}$
12.	0,55	$6,67 \times 10^{-2}$
13.	0,60	$7,18 \times 10^{-3}$
14.	0,65	$7,87 \times 10^{-3}$
15.	0,70	$8,79 \times 10^{-3}$
16.	0,75	$1,01 \times 10^{-2}$
17.	0,80	$1,19 \times 10^{-2}$
18.	0,85	$1,46 \times 10^{-2}$
19.	0,90	$1,92 \times 10^{-2}$
20.	0,95	$2,84x10^{-2}$
21.	1,00	-

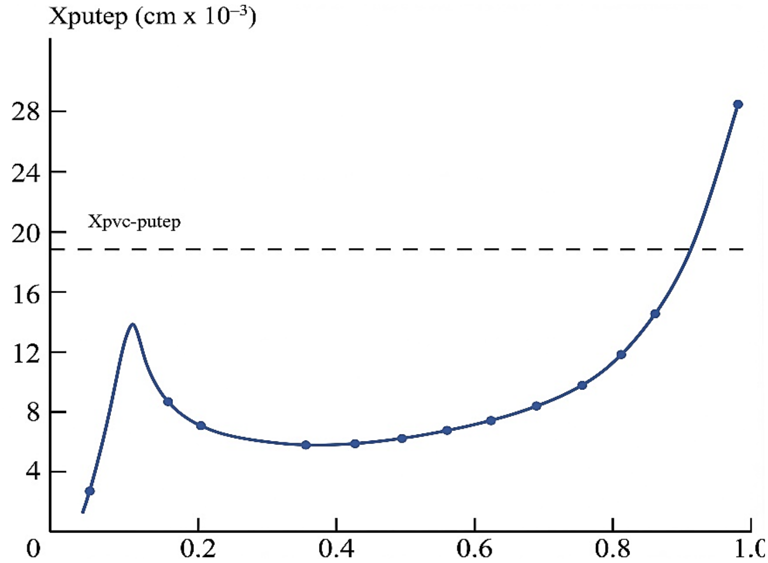


Figure 2. Theoretical phase diagram of the PVC-PUTEP system.

It is evident that PUTEP exhibits significantly higher solubility in PVC compared to PVC itself, and when the PUTEP content remains below 10%, complete compatibility within the system is expected. It follows from the calculation that the so-called asymmetric case is observed with two critical dissolution temperatures - lower and upper.

Result

1. The calculation method shows that BR and HDPE polymers are practically incompatible. The variational approach was employed to estimate the concentration (0.08 by mass) at which complete association can occur.
2. It is evident that PUTEP exhibits significantly higher solubility in PVC compared to PVC itself, and when the PUTEP content remains below 10%, complete compatibility within the system is expected.
3. The evaluation of the generalized desirability functions indicated that increasing the PVC concentration from 5 to 10 wt.% resulted in a rise in the desirability value (D) from 0.067 to 0.078.

CONCLUSİON

The compatibility of polymer systems is essential for influencing the mechanical, thermal, and chemical properties of blended materials. This study investigates the compatibility behavior of selected polymer blends and evaluates the chemical properties of their mixtures. Compatibility was assessed through physicochemical characterization techniques including Fourier Transform Infrared Spectroscopy (FTIR), Differential Scanning Calorimetry (DSC), and Scanning Electron Microscopy (SEM) to identify intermolecular interactions, glass transition behavior, and morphological features. The presence of single or shifted glass transition temperatures and specific spectral changes were used as indicators of miscibility and interaction between polymer phases. Chemical stability, functional group interactions, and resistance to environmental factors were also examined. The findings indicate that intermolecular interactions, including hydrogen bonding and dipole–dipole forces, play a crucial role in determining the compatibility of the polymer blend. Understanding these interactions provides valuable insight into tailoring polymer mixtures with enhanced performance for applications in coatings, packaging, biomedical materials, and advanced composites.

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