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Investigation of the Compatibility of Binary and Ternary Polymer Systems

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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. These groups provide a broad range of enhanced properties – including improved adhesive performance, higher tensile and tear strength, and greater resistance to thermal and chemical degradation – primarily due to intermolecular polar interactions among macromolecules. 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 the retention times of sorbents, the solubility parameter value at 25°C was calculated and subsequently compared with the value obtained by the TDT method at the same temperature. It was observed that the discrepancy between the solubility parameter values determined by the two methods increased with increasing polymer polarity. This phenomenon is attributed to the fundamentally contrasting states of the investigated 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

INTRODUCTION

Currently, at the industrial level, the challenge of producing polymeric materials with properties suitable for modern applications is addressed through two primary approaches: the synthesis of new polymer varieties and the development of polymer composites in which the characteristics of the individual components complement each other in an additive manner [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

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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 accomplish this objective, it is essential to evaluate the compatibility and interaction parameters of polymer blends. 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 of the properties of newly developed polymer compositions, along with the optimization of their processing conditions using existing industrial equipment, enables the identification of thermodynamic compatibility patterns among various commercially produced polyolefin and heterochain polymers. This approach facilitates the production of composite materials with substantially enhanced performance characteristics. The integration of diverse polyfunctional components possessing optimal properties underscores 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 composite performance, achieving marked improvements in heat resistance under air and vacuum conditions as well as enhanced anticorrosive behavior [20–24]. In this context, oil–polymer resin SPP has been introduced into composite systems for manufacturing products designed to operate under severe mechanical wear, elevated temperatures, high pressures, stresses, and deformation conditions [25–29].

Accordingly, the development of advanced polymer compositions that simplify production and processing technologies while lowering manufacturing costs and delivering required property combinations remains highly important under current technological conditions [30–41]. This need arises primarily from the insufficient development of the theoretical framework governing multicomponent polymer systems and the absence of a comprehensive scientific basis for such 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 [47–68].

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.

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 (Figure 1).

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).

Figure 1. chromatography-mass spectrometry.

m $\Delta F_{см}$, $\Delta H_{см}$, $\Delta S_{см}$

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:

$$\Delta F_{см} = \Delta H_{см} - T\Delta S_{см} = 0 .$$

From here:

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

where $T_{кр}$ – 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:

$Qp = k(-Ei_n + E_{eb} + 2E_{in-eb.}$

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:

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

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 Butyl Rubber–Low-Density Polyethylene (BR–LDPE) System at 250°C

To determine the solubility parameters of each component in the blend, tabulated values of molar attraction constants corresponding to different low-molecular-weight chemical groups were utilized. The densities of the polymers employed in this study were experimentally evaluated. The obtained values were: for butyl rubber (BR) at 250°C, ρ = 0.92 × 10^3 kg/m³, and for low-density polyethylene (LDPE), ρ = 0.96 × 10^3 kg/m³. In these calculations, the influence of terminal functional groups and the molecular weight distribution of the polymer systems was not considered.

$CH_3|[–CH_2–C–]_n – [–CH_2–C=CH–CH_2]_m|CH_3\ CH_3$

Molar constants of attraction of isoprene:

$\sum F = 133.2 + 93 + 214 + 28 = 415$

$M = 5 \times 12 + 8 \times 1 = 68$

Molar constants of attraction of isobutylene:

$\sum F – CH_2–CH_3$ \ /
C
/ \

$\sum F = 133 + 2 \times 214 – 93 = 468$

$M = 12 \times 4 + 5 \times 1 = 53$

Then the solubility parameter of butyl rubber:

$$\delta_{бк} = 0.92/0.95\,\frac{468}{53} + 0.05\,\frac{415}{68} - 8.0$$

Solubility parameter HDPE:

$\sum F = 133$

$M = 1 \times 12 + 2 \times 1 = 14$

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 (cm³/moll), usually taken to be 100 cm³/mol. For this value of Vp and a temperature of 25°C, the Hildebrand equation takes the form: $\delta_{бк} = 0.96\,\frac{133}{14} = 9.12$

In this case = $(\delta_{A} - \delta_{B)}$

X_{BR-} 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 can be determined from the actual degree of polymerization (X), provided that the molar volume of the polymer's repeating unit is known, using the following equation:

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

A fairly good approximation is obtained from the relation:

XA = MA/100

where MA is the molecular weight of the polymer.

Thus, we have:

$XBR = \frac{390000}{100} = 3900$

$X\ HDPE = \frac{320000}{100} = 3200$

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

(Xbr-hdpe) = 1/2 x 1/39001/2 + 1/32001/2 = 7.18 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.

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

F_{br}	**$(X_{br-hdpe})_{sp}$**	**Fbr**	**$(X_{br-hdpe})_{sp}$**
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	–	–

As shown in Table 1, decreasing the concentration of any component in the mixture leads to an increase in the spindle solubility parameter. Moreover, HDPE exhibits significantly better solubility in BR than BR does in high-density polyethylene. The concentrations corresponding to the absolute dilution of the original polymers were determined using the variation method. These values were found to be 0.08 wt% for HDPE and 0.06 wt% for BR.

The calculations indicate that the BR–HDPE system is essentially incompatible. Therefore, to identify the critical point of this system on the phase diagram, it is necessary to determine the Gibbs free energy of mixing (G_{sm}) for the components involved. 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 = 25°C, the results of which are presented in Table 2.

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 data obtained, 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 data obtained show 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%.

During the process, the compatibility of the components is strongly influenced by their polarity. Based on this consideration, we evaluated the compatibility parameters for the polar polymer system comprising PVC and PUTPE and compared the results with those of the non-polar BR–HDPE system.

CALCULATION OF THE COMPATIBILITY OF A THERMOELASTOPLASTICIZED POLYVINYL CHLORIDE–POLYURETHANE (PVC–PUTEP) SYSTEM AT 25°C

To determine the solubility parameters of each component in the blend, tabulated values of the molar attraction constants for different molecular/homogeneous groups (MHGs) were utilized, considering the density of PVC at 25°C ($\rho_{25} = 1.41 \times 10^3$ kg/m^3). The calculations were performed both by accounting for the distribution of terminal groups along with the molecular weight distribution and by neglecting the molecular weight distribution, to evaluate its influence on the compatibility assessment.

Single link PVX – [–CH_2 – CHCl –] n

Molar constants of attraction:

–CH_2 – –CH = Cl –

∑F = 133 + 28 + 270 = 431

M = 12.2+ 1.3 + 35.5 = 62.5

Then the PVX solubility parameter will be:

Δ_{pvx} = 1.41 x 431/62.5 = 9.72

The solubility parameter of PUTEP, considering the density of PUTEP at 25°C ($\rho 25 = 1.1 \text{ x } 103 \text{ kg/m}^3$). Single link PUTEP:

$[-(CH_2)_6 - OCOHN - (CH_2)_4 -]_n$

$-(CH_2)_6- \quad -COO- \quad -NH- \quad -(CH_2)_4$

$\sum F = 133.6 + 310 + 180 + 133.4 = 757$

$M = 12.11 + 1.21 + 16.2 + 14 = 199$

$\Delta_{putep} = 1.10 + 1820 + 199 = 10.06$

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 represents the gas constant, equal to 1.987 kcal mol^{-1} deg^{-1}; T denotes the temperature in Kelvin; and V_n refers to the reference molar volume (cm^3/mol), typically assumed to be 100 cm^3/mol. Therefore, when T = 25°C (298 K), the equation can be written in the following 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:

$Xcr = 1/2[1/X1/2A + 1/XB1/2]2$

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:

$XA = (V / Vp) \, x$

A fairly good approximation is obtained from the relation:

$XA = MA/100$

where MA is the molecular weight of the polymer. Thus, we have:

$Xpvx = \frac{90000}{100} = 900$

$Xputep = \frac{18000}{100} = 180$

Отсюда по уравнению для расчета критического значения:

$(X_{pvc\text{–}putep})$ critical temperature $= 1/2 \text{ x } 1/X_A \, (\Phi_A / G_п) + 1/X_в \, (\Phi B)_{sp}$

The results obtained are shown in Table 3.

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 × 10–3
3.	0.10	1.40 × 10–2
4.	0.15	8.64 × 10–3
5.	0.20	6.97 × 10–3
6.	0.25	6.25 × 10–3
7.	0.30	5.93 × 10–3
8.	0.35	5.82 × 10–3
9.	0.40	5.86 × 10–3
10.	0.45	6.02 × 10–3
11.	0.50	6.29 × 10–3
12.	0.55	6.67 × 10–2
13.	0.60	7.18 × 10–3
14.	0.65	7.87 × 10–3
15.	0.70	8.79 × 10–3
16.	0.75	1.01 × 10–2
17.	0.80	1.19 × 10–2
18.	0.85	1.46 × 10–2
19.	0.90	1.92 × 10–2
20.	0.95	2.84 × 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 × 10–3
3.	0.10	1.40 × 10–2
4.	0.15	8.64 × 10–3
5.	0.20	6.97 × 10–3
6.	0.25	6.25 × 10–3
7.	0.30	5.93 × 10–3
8.	0.35	5.82 × 10–3
9.	0.40	5.86 × 10–3
10.	0.45	6.02 × 10–3
11.	0.50	6.29 × 10–3
12.	0.55	6.67 × 10–2
13.	0.60	7.18 × 10–3
14.	0.65	7.87 × 10–3
15.	0.70	8.79 × 10–3
16.	0.75	1.01 × 10–2
17.	0.80	1.19 × 10–2
18.	0.85	1.46 × 10–2
19.	0.90	1.92 × 10–2
20.	0.95	2.84 × 10–2
21.	1.00	–

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 (ΦA)sp + 1/XB(ΦB)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 Table 4 and Figure 2.

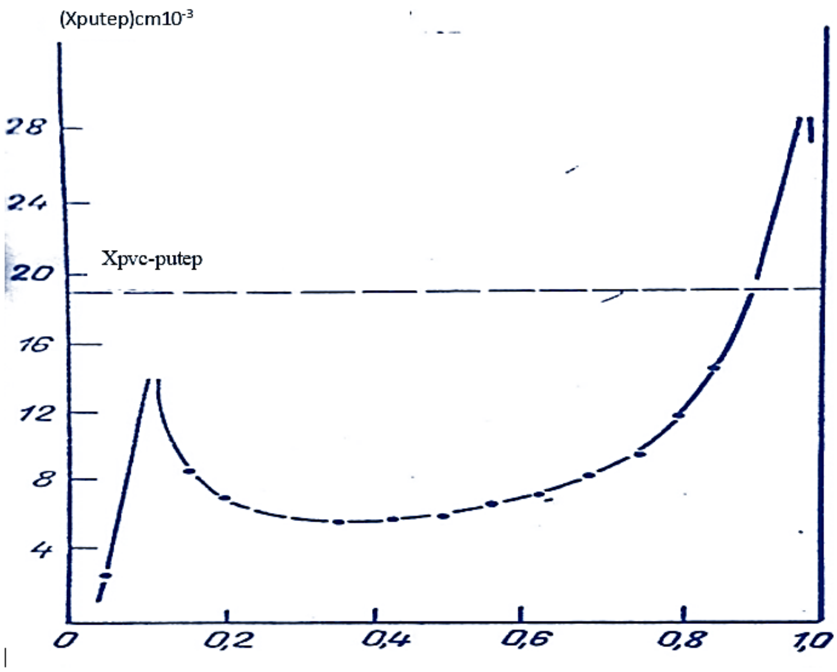

Figure 2. Theoretical phase diagram of the PVC-PUTEP system.

As can be seen, PUTEP dissolves in PVC much better than PVC, and at a content of less than 10% PUTEP, full system compatibility should be observed. It follows from the calculation that the so-called asymmetric case is observed with two critical dissolution temperatures – lower and upper.

RESULT

The calculation method shows that BR and HDPE polymers are practically incompatible. The variational method was used to determine the concentration at which absolute combination is possible (0.08; mass).

1. It is shown that PUTP dissolves in PVC much better than PVC. When the content is less than 10% of the mass. PUTEP shows full compatibility with polymers.
2. An analysis of the generalized desirability functions showed that an increase in the PVC content from 5–10 wt. Leads to an increase in D from 0.067 to 0.078.

CONCLUSİON

The present investigation systematically evaluated the compatibility of binary and ternary polymer systems through physicochemical, thermal, and structural analyses. The results demonstrate that polymer compatibility strongly depends on intermolecular interactions, compositional ratios, and processing conditions. Binary blends exhibited partial to complete miscibility when favorable interactions, such as hydrogen bonding or dipole–dipole interactions, were present, whereas immiscible systems showed phase separation and reduced homogeneity. In ternary systems, the incorporation of a third component significantly influenced phase behavior, often enhancing compatibility by acting as a compatibilizing agent and improving interfacial adhesion. Thermal analysis indicated modifications in glass transition temperatures, confirming molecular-level interactions in compatible systems. Overall, the study highlights that careful selection of polymer components and optimization of blend ratios can tailor material properties effectively. These findings provide valuable insight into designing advanced polymeric materials with improved mechanical, thermal, and functional performance for industrial and engineering applications.

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