

Compatibility Studies of Partially Compatible Blends Through Glass Transition Temperature and Melting Point Depression Analysis

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ABSTRACT: This work deals with a compatibility study of blends based on poly(vinylidene fluoride) (PVDF) by means of the determination of the polymer–polymer interaction parameter through two different and separated methods. Firstly, the well-known Flory–Huggins interaction parameter was determined from melting point depression analysis and subsequently from T_g measurements by applying an expression proposed by Kim and Burns. © 1998 John Wiley & Sons, Inc. *J Appl Polym Sci* **67**: 997–1004, 1998

Key words: polymer–polymer interaction parameter; compatible blends; poly(vinylidene fluoride); melting point depression; glass transition temperature

INTRODUCTION

At present, blending or alloying polymers constitutes one of the most attractive approaches to obtain new polymeric materials with specific characteristics.¹ Due to their importance on the field of polymer applications, special attention is given to those blends in which the component polymers are believed to be mixed on the molecular level, yielding miscible mixtures; miscibility is, however, known to be a rare phenomenon in practice. From an exclusively thermodynamic point of view, the blends may be classified as miscible, partially miscible, and immiscible. Miscibility is linked to the thermodynamic concept of homogeneity, to the existence of a single phase, and to property isotropy. In other words, a miscible blend is the genuine dissolution of the one polymer in the other. Hence, the intrinsic state of a blend is similar to that occurring between two miscible organic liquids.

From a technological stance, however, the ap-

proach to miscibility is totally different. It is sufficient for a material to behave as if constituted by a single phase for it to be categorized as “compatible”; although, thermodynamically speaking, it is actually an immiscible or partially miscible blend, yet with a high degree of homogeneity and phase dispersion. Hence, for practical purposes, it is indispensable that there exists a certain compatibility when polymer blends with reasonable technological properties are at stake.

As a consequence of the discovery of the phenomenon of piezoelectricity in quartz, a great number of inorganic materials have been developed with extraordinary pyro-piezoelectric features, among which the ceramics of the barium titanate type deserve to be specially highlighted. All of these, however, are pulverulent, dusty materials, hard and stiff, with bad mechanical properties and difficult to obtain for mass production, apart from resisting moulding into complex shapes and their high price.

On the other hand and due to the spectacular progress made in electronics in the last years, there is a growing demand for materials with pyro-piezoelectric properties. These latter materials acquire special relevance in the field of

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physical sensors or transducers, as they themselves are capable of transforming mechanical stress into an electric output and vice versa. This property is highly appreciated in quite different fields, such as acoustics, ultrasound, medicine, etc.; but most especially in robotics,² where these materials find their most genuine application. With the aim of overcoming the inconvenience and limitations of ceramics, a study was launched, examining the possibility of synthesizing polymeric materials with pyro-piezoelectric properties.³⁻⁸ Among these poly(vinylidene fluoride) (PVDF) has become the polymer that generates the greatest scientific interest. Based on PVDF a series of new materials have been developed, either through copolymerization or else by blending PVDF with polymethacrylates or fluorinated polymers.⁹

At this stage, it has to be mentioned that one of the major drawbacks in obtaining new pyro-piezoelectric materials based on PVDF blends is its low compatibility with most other polymers. When a potentially pyro-piezoelectric blend is incompatible, it contains nonelectroactive phases and interfaces, which constitute barriers to the transfer and movement of the electrical charges. Hence, the synthesis of new pyro-piezoelectric materials is conditioned to the existence of a certain compatibility among them.

In a previous article,¹⁰ we studied the compatibility of different blends based on PVDF, analyzing the changes observed in the glass transition temperature from dielectric and calorimetric data. When T_g was analyzed by differential scanning calorimetry (DSC), all samples displayed only one change of the heat capacity corresponding to the glass transition temperature. This fact, according to the single T_g criterion, indicates that it is possible to consider these blends as miscible. With regard to the dielectric data, all samples showed the T_g s corresponding to the homopolymers, but significantly shifted with respect to the unblended homopolymers. We explained this disagreement, concluding that a given system may not be molecularly homogeneous under a particular set of conditions and that individual molecules of each component may cluster in very small domains; i.e., microheterogeneities may appear. Furthermore, if different techniques are responsive over different ranges, one may be led to different conclusions concerning the compatibility of the blend; so the blends themselves were considered as partially miscible.

In this work, which further pursues previous

research,^{11,12} the objective is to prepare polymer blends, probably with pyro-piezoelectric characteristics, based on PVDF, poly(vinyl acetate) (PVA), and poly(methyl methacrylate) (PMMA) and determining experimentally their glass transition temperature and melting point depression to study the polymer-polymer interaction parameter.

EXPERIMENTAL

Materials

All the polymers used were commercial products and were used as received. The PVDF was Solef 6010 supplied by Solvay (Germany) with $M_n = 64,000$ and $\rho = 1.78 \text{ g/cm}^3$; PVA was Mowilith 50, a Hoechst (Germany) product, $M_n = 99,110$ and $\rho = 1.17 \text{ g/cm}^3$; and the PMMA was supplied by Repsol S.A. (Madrid, Spain) with $M_n = 63,770$ and $\rho = 1.18 \text{ g/cm}^3$.

Blending

Blends were prepared in a Brabender Plasticorder using a thermoplastic mixing chamber type W60 preheated at 180°C; rotor speed was set at 60 rpm, and 10 min of mixing were enough to get uniform dispersion of the components.

Methods

Isothermal crystallization from the melt was studied using a Perkin-Elmer model DSC 7 differential scanning calorimeter operating in an N_2 atmosphere. The following procedure was employed: the samples were kept for 5 min at 493 K to destroy their thermal history and then quenched (350 K min) to their crystallization temperature T_c . The melting temperature T_m of each sample after isothermal crystallization at T_c was calculated by heating the sample directly from T_c to T_m at a heating rate of 5 K min.

A mettler TA4000 differential scanning calorimeter was used for calorimetric measurements. The heating rate was 10°C/min; and before recording DSC thermograms, samples were quenched to low temperature from the melt. The midpoint of the slope change of the heat capacity plot was taken as the glass transition temperature. The DSC thermograms for every samples

were carried out three times. The T_g data employed was the average value.

Dynamic mechanical properties were analyzed with a DMTA Polymer Laboratory. Frequencies chosen were 3 and 10 Hz, and the temperature range was between -150 and $+150^\circ\text{C}$.

RESULTS AND DISCUSSION

Blend Compositions

In order to obtain new materials with desirable properties, the usual procedure is to prepare, at random, more or less complicated mixtures of the different components and to select the most suitable empirically. However, we have used the classical Scheffé Simplex model,¹³ a statistical design, to plan the compositions of the three polymers in the blends. The following condition is fulfilled: $\sum_i Z_i = 1$, where Z_1 , Z_2 , and Z_3 are the weight percentages of PVDF, PVA, and PMMA, respectively.

The objective is to obtain an empirical equation that satisfactorily describes the properties of the mixture over the whole area which is used. For this purpose, Scheffé proposed the study of mixtures whose compositions are distributed symmetrically in a "simplex" lattice over the experimental range. This kind of design contains a number of points equal to the coefficients in the corresponding polynomial, which allows the equation to be solved directly. The equation, also named the equation of response surfaces, is a polynomial^{14,15} and corresponds to the development of Taylor serial functions.

In our case, we have considered that only blends with PMMA or PVA content, which make up less than 60% in the blend, show pyro-piezoelectric behavior, so we applied the Scheffé model

and blends were prepared according to the following plan.

With the results of experiments 1–6 and using the Software Nemrod¹⁶ coefficients for a quadratic model,

$$y = b_1Z_1 + b_2Z_2 + b_3Z_3 + b_{12}Z_{12} + b_{13}Z_1Z_3 + b_{23}Z_2Z_3 \quad (1)$$

were calculated; while experiment 7 was used as test in the model.

Polymer–Polymer Interaction Parameter Calculated from Melting Point Depression

According to the theory of Scott,¹⁷ and modified by Nishi and Wang,¹⁸ the melting depression point occurs when there exists thermodynamic blending of a crystalline and an amorphous polymer, by analogy with crystalline polymer–polymer diluent systems, and can be described by the following equation:

$$1/T_m^* - 1/T_m^0 = -[(RV_{2u}/\Delta H_{2u}V_{1u})]\chi_{12}\Phi_1^2 \quad (2)$$

where the subscript 1 identifies the amorphous polymer, and the subscript 2 identifies the crystalline polymer. Φ is the volume fraction, V_u is the molar volume of repetitive units, ΔH_{2u} refers to the melting enthalpy per mole of the repetitive unit, T_m^0 is the equilibrium melting temperature of the unblended crystalline polymer, T_m^* represents the equilibrium melting temperature of the crystalline polymer in the blend, and χ_{12} stands for the polymer interaction parameter.

The plots of $(1/T_m^* - 1/T_m^0)$ against Φ_1^2 give rise to straight line, from whose slope, the polymer–polymer interaction parameter may be calculated.

Exp. No.	Blends	Composition (wt %) PVDF–PVA–PMMA	Variables		
			Z_1	Z_2	Z_3
1	PVDF	100/0/0	1	0	0
2	FA64	60/40/0	0	1	0
3	FM64	60/0/40	0	0	1
4	FAM622	60/20/20	0	0.5	0.5
5	FA82	80/20/0	0.5	0.5	0
6	FM82	80/0/20	0.5	0	0.5
7	FAM811	80/10/10	0.5	0.25	0.25

Table I Compositions and Equilibrium Melting Point (Experimental and Calculated Applying the Scheffé Model) for the Different Blends

Sample	Composition (Φ_V) PVDF–PVA–PMMA	Equilibrium Melting Point (K)
PVDF	100/0/0	450.9
FA91 ^a	85.5/14.5/0	450.0
FA82	72.4/27.6/0	449.0
FA73 ^a	60.5/39.5/0	447.9
FA64	49.35/50.35/0	446.8
FM91 ^a	85.7/0/14.3	449.5
FM82	72.6/0/27.4	448.1
FM73 ^a	60.7/0/39.3	446.9
FM64	49.9/0/50.1	445.7
FAM622	49.8/25.2/25.0	449.2
FAM811	72.5/13.8/13.7	450.1

^a Calculated.

A large positive χ_{12} indicates unfavorable interaction and that polymer mixtures are immiscible. A low value indicates little interaction (partially miscible blend); and a large negative value indicates a rather strong specific interaction, i.e., polymer miscibility.

Isothermal crystallization¹⁹ was carried out for all the blends proposed in the model; and, from the results, the equilibrium melting temperatures were calculated by the Hoffman–Weeks²⁰ graphs of T_m versus T_c , where T_m^* is the intercept node of T_m with the straight line $T_m = T_c$, according to the following expression:

$$T_m = (1/\eta)T_c + (1 - 1/\eta)T_m^* \quad (3)$$

$1/\eta$ is the so-called morphological parameter, whose value is practically constant and independent of the blend composition.²¹

Applying the Scheffé model proposed, the following equation for T_m^* was obtained:

$$T_m^* = 450.9 Z_1 + 446.8 Z_2 + 445.7 Z_3 + 0.6 Z_1 Z_2 - 0.81 Z_1 Z_2 + 11.8 Z_2 Z_3 \quad (4)$$

So, in Table I, we show the experimental, besides the calculated values, using the above equation.

The graphic representation of eq. (2), plotting $1/T_m^* - 1/T_m^0$ versus Φ_1^2 (where $\Delta H_{2u} = 1.6$ Kcal mol⁻¹; $V_{2u} = 36$ cm³ mol⁻¹; $V_{1u}^{PVA} = 75$ cm³ mol⁻¹; and $V_{1u}^{PMMA} = 85$ cm³ mol) allows the determina-

tion of the polymer–polymer χ_{12} interaction parameters from the slopes of the straight lines thus obtained.

The results obtained are shown in Table II, together with the Y-intercepts, which are practically zero as no entropic contribution to melting point depression exists. In both cases, whether the amorphous polymer in blend is PVA or whether it is PMMA, the polymer–polymer interaction parameters χ_{12} are negative.

Polymer–Polymer Interaction Parameter Calculated from Glass Transition Temperature Measurements

It is well-known that a miscible blend shows a single T_g intermediate between those of the components in isolation; whereas, for immiscible blends, T_g s of each component are detected.^{22,23} However, in our case, as above-mentioned, the studied blends show a single T_g when determined by DSC and the T_g s corresponding to each of the components when determined by dielectric or dynamic mechanical measurements.

Theoretically, when two polymers are “totally” miscible, the T_g of the blend can be estimated empirically from the individual components T_g s and their weight percentage in the blend, applying different theories, such as the Fox eq. (24), as follows:

$$(1/T_g^b) = w_1/T_{g1} + w_2/T_{g2} \quad (5)$$

or the Wood eq. (25), as follows:

$$T_g^b = w_1 T_{g1} + w_2 T_{g2} \quad (6)$$

where w_1 and w_2 are the weight percentages of polymers 1 and 2, respectively; and T_{g1} and T_{g2} the glass transition temperatures of each component in isolation. T_g^b stands for the glass transition of the blend.

But, when two polymers are partially miscible (which here are designated as 1 and 2) are blended, there occurs stabilization into two phases at the end of the mixing process, one being rich in polymer 1 (phase 1), and the other dominated by polymer 2 (phase 2). Each of these phases may be considered as totally miscible system, as no further phase separation occurs once the equilibrium has been reached; hence, Fox’s or

Wood's equations are applicable to each of them, so as to quantify the portion of one and the other component in blend.

Now then, the Fox's equation can be rearranged to

$$w_{11} = [T_{g1}(T_{g1}^b - T_{g2})/T_{g1}^b(T_{g1} - T_{g2})] \quad (7)$$

Similarly, the Wood's equation can be rewritten as

$$w_{11} = (T_{g1}^b - T_{g2})/(T_{g1} - T_{g2}) \quad (8)$$

where w_{11} is the weight fraction of polymer 1 in phase 1, $w_{21} = 1 - w_{11}$ is the weight fraction of polymer 2 in phase 1, and T_{g1}^b is the glass transition temperature of phase 1 in which polymer 1 is predominant.

Analogously, the polymer 1 portion can be determined for phase 2, rewriting expressions (7) or (8), respectively, as

$$w_{12} = [T_{g1}(T_{g2}^b - T_{g2})/T_{g2}^b(T_{g1} - T_{g2})] \quad (9)$$

$$w_{12} = (T_{g1}^b - T_{g2})/(T_{g1} - T_{g2}) \quad (10)$$

where w_{12} is the weight fraction of polymer 1 in phase 2, rich in polymer 2; and T_{g2}^b stands for the glass transition temperature of phase 2.

Once the weight fractions have been determined, the volume fractions can be calculated via the following ratios:

$$\Phi_{11} = [(w_{11}/\rho_1)/(w_{11}/\rho_1 + w_{21}/\rho_2)];$$

$$\Phi_{21} = 1 - \Phi_{11} \quad (11)$$

where Φ_{11} and Φ_{12} are the volume fractions of polymers 1 and 2, respectively, in phase 1; and ρ_1 and ρ_2 represent the respective polymer densities.

The Flory-Huggins polymer-polymer interaction parameter χ_{12} is determined by applying the expression developed by Kim and Burns,²⁶ applicable to partially compatible systems in the absence of solvents and assuming that the equilib-

Table II Interaction Parameters and Intercepts of PVDF-PVA and PVDF-PMMA Systems at 433 K

System	χ_{12}	Intercept $\times 10^6$
PVDF-PVA	-0.164	2.22
PVDF-PMMA	-0.153	6.61

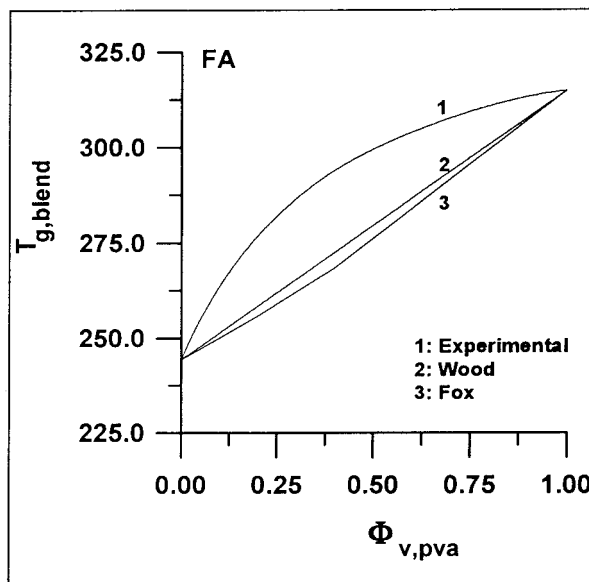


Figure 1 Glass transition temperature behavior for PVDF-PVA systems. Experimental data were obtained through DSC analysis.

rium state has been accomplished for the following system:

$$\chi_{12} = \frac{(\Phi_{11}^2 - \Phi_{12}^2)[m_2 \ln \Phi_{12}/\Phi_{11} + (m_1 - m_2)(\Phi_{21} - \Phi_{22})] + (\Phi_{21}^2 - \Phi_{22}^2)[m_1 \ln \Phi_{22}/\Phi_{21} + (m_2 - m_1)(\Phi_{11} - \Phi_{12})]}{2m_1m_2(\Phi_{11}^2 - \Phi_{12}^2)(\Phi_{21}^2 - \Phi_{22}^2)} \quad (12)$$

m_i is essentially the degree of polymerization, relating the molar volumes V_1 and V_2 of the polymers to a fictitious molar volume V_0 of one submolecule of polymer, as follows:

$$m_1 = V_1/V_0; \quad V_1 = M_{n1}/\rho_1 \quad (13)$$

$$m_2 = V_2/V_0; \quad V_2 = M_{n2}/\rho_2 \quad (14)$$

where M_n is the number-average molecular weight, and ρ is the density of polymer.

Taking into account some mathematical considerations, it is possible to find the critical condition, as follows:

$$(\chi_{12})_c = \frac{1}{2}(m_1^{-1/2} + m_2^{-1/2}) \quad (15)$$

The choice of a lattice site volume can be rather arbitrary; but once a site volume has been chosen for one of the components, it must be the same for the other component. In our case, the repeat

Table III Polymer–Polymer Interaction Parameters and Volume Fractions Obtained by the Equation of Kim and Burns

Blend	3 Hz			10 Hz		
	Φ_{12}	Φ_{21}	$\chi_{12} \times 10^3$	Φ_{12}	Φ_{21}	$\chi_{12} \times 10^3$
FA82	0.364	0.120	1.309	0.347	0.111	1.319
FA64	0.599	0.517	2.025	0.631	0.078	1.933
FM82	0.180	0.156	1.640	0.156	0.170	1.651
FM64	0.679	0.093	2.201	0.792	0.101	2.655

unit of the amorphous polymer (PVA or PMMA) has been chosen as a lattice site volume.

In order to apply the above equation proposed by Kim and Burns, we have considered the dynamic–mechanical²⁷ results since the T_g s corresponding to each of the components of the blend are displayed.

Now then, in order to get the best fit (Fox or Wood equation) to our systems, we have plotted the calorimetric²⁷ and theoretical data. In Figures 1 and 2, we show the glass transition temperature behavior for PVDF–PVA and PVDF–PMMA, respectively.

So, for PVDF–PVA blends, we have chosen the Wood's equation only to calculate the w_{11} weight fraction and the Fox's equation for PVDF–PMMA systems.

Table III compiles the polymer–polymer inter-

action parameters and volume fractions obtained from the method proposed.

The miscibility of one polymer in the other was examined by plotting the volume fractions of each polymer in the other phase as a function of the respective amorphous polymer content. Thus, Figures 3 and 4 show, respectively, the PVDF–PMMA and PVDF–PVA systems. Φ_{21} refers to the PVDF portion present in the PMMA- or PVA-rich phase, whereas Φ_{12} shows the PMMA or PVA content in the phase rich in PVDF.

PMMA or PVA are observed to dissolve more easily in the PVDF-rich phase than vice versa as Φ_{12} values are considerably greater than the Φ_{21} ones, although the latter are not zero, as it described for totally incompatible blends.²⁸ In short, the amount of PMMA or PVA dissolved in the PVDF-rich phase increases considerably when increasing the amorphous polymer portion. As regards the miscibility of PVDF in the PMMA or PVA-dominated phase, the level is not very high and diminishes with the amorphous polymer content.

For PVDF–PVA system and using $m_1 = 1152.5$ and $m_2 = 489.2$, we can also determine the critical value of χ_{12} ; it has been found to be $(\chi_{12})_c = 0.00279$. Similarly, for PVDF–PMMA systems with $m_1 = 637.7$ and $m_2 = 424.2$, $(\chi_{12})_c = 0.00389$. It can be surmised that if $\chi_{12} < (\chi_{12})_c$ is observed, then the polymers are miscible with each other; and there will be no phase separation. If $\chi_{12} > (\chi_{12})_c$ is observed in blends, phase separation will occur.²⁹ For both studied systems, the value of χ_{12} is greater than the value of $(\chi_{12})_c$.

CONCLUSIONS

Interpretation of these findings allows conclusion of the following.

1. The values corresponding to χ_{12} , when ob-

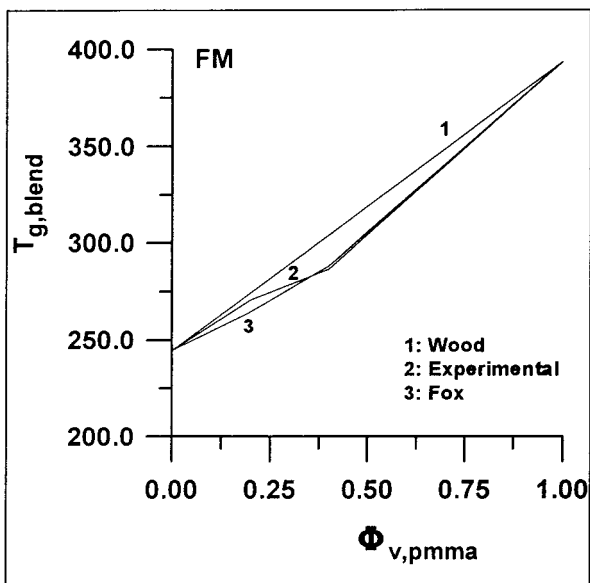


Figure 2 Glass transition temperature behavior for PVDF–PMMA systems. Experimental data were obtained through DSC analysis.

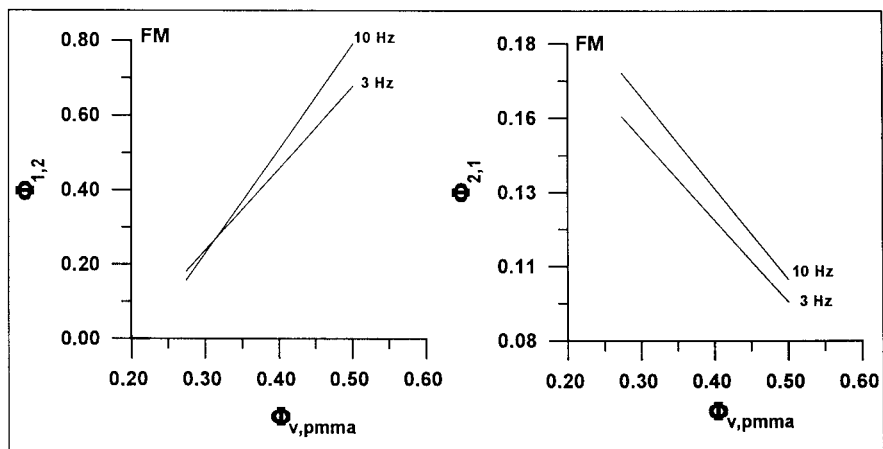


Figure 3 Volume fractions of each polymer in the other phase as a function of PMMA content.

tained from melting point depression analysis and for both systems, are low and negative.

- The disagreement with DSC, on the one hand, and mechanical and dielectric measurements, on the other hand, made us think that these are actually partially miscible blends; so we decided to apply the equation proposed by Kim and Burns (the T_g s values employed were obtained from dynamic mechanical measurements). In this case, χ_{12} has positive and very low values.

This fact indicates that there exists a moderate interaction between the polymers that constitute the systems. This interaction is enough for get a high degree of

homogeneity and phase dispersion; so, the material can be considered as if constituted by a single phase but not a blend on the molecular level. This assumption is corroborated because both systems fulfill the condition concerns $(\chi_{12})_c$; that is to say, as $\chi_{12} < (\chi_{12})_c$, then the polymers have some affinity with each other, and there will be no phase separation.

- The weight fraction of PMMA or PVA dissolved in PVDF-rich phase is higher that the weight fraction of PVDF component dissolved in the PMMA or PVA-rich phase. These results suggest that the PMMA or PVA dissolves more in the PVDF-rich phase than does the PVDF in the PMMA- or PVA-rich phase.

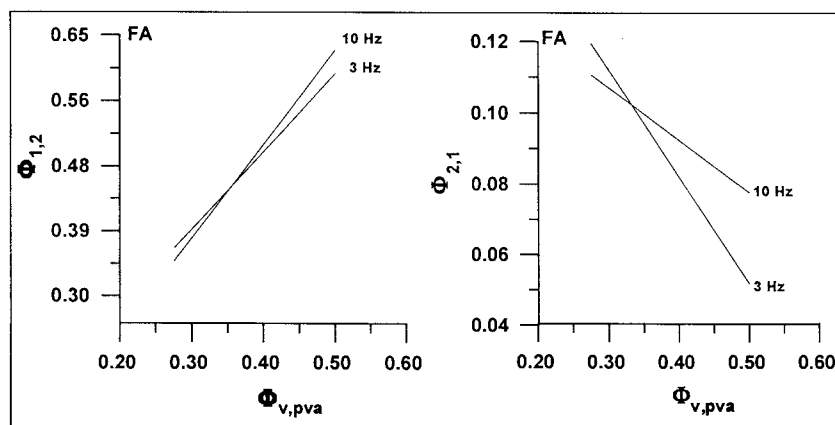


Figure 4 Volume fractions of each polymer in the other phase as a function of PVA content.

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