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COMPATIBILITY STUDIES ON POLYBLENDS OF BUTYL RUBBER WITH PPG AND PEG USING ULTRASONIC TECHNIQUES

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Abstract

This study investigates the compatibility of butyl rubber (BR) with polypropylene glycol (PPG) and polyethylene glycol (PEG) using ultrasonic measurements at 303 K. Ultrasonic parameters—adiabatic compressibility, free volume, and internal pressure—were used to evaluate intermolecular interactions in BR–PPG and BR–PEG blends at 0.5% and 1% compositions. Decreasing compressibility and free volume, along with increasing internal pressure, indicate enhanced molecular interactions and improved packing with rising polyether content. Comparative analysis shows that PEG exhibits greater compatibility with BR than PPG, likely due to its higher polarity. Ultrasonic evidence confirms homogeneous mixing without phase separation across all compositions. Overall, the study demonstrates the effectiveness of ultrasonic techniques in assessing polymer blend miscibility and provides insights useful for designing high-performance BR-based materials.

Keywords: Ultrasonic Techniques, Butyl Rubber (BR), Polymer Blend Compatibility, Polypropylene Glycol (PPG), Miscibility Analysis, Polyethylene Glycol (PEG), Adiabatic Compressibility, Liquid Mixtures.

1. Introduction

Polymer polyblends are designed and developed by incorporating complimentary properties of different macromolecular constituents to yield materials with custom mechanical, thermal, and chemical behaviour [1]. Butyl rubber, also known as isobutylene isoprene rubber, or IIR/BR, is one of the most popular elastomers for applications requiring low gas permeability, damping, and chemical resistance [2]. By combining with polar polyethers like polypropylene glycol (PPG) and polyethylene glycol (PEG), the natural properties of butyl rubber can be further modified. The exploration of the compatibility of such polyblends is also essential since morphology, phase continuity, and, therefore service performance in a broad variety of applications are determined by miscibility and interfacial interactions, including vibration isolation, sealants, and barrier

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coats [3].

Ultrasonic methods provide a very sensitive, non-destructive probe of molecular interaction, free volume change and elastic response of polymers and their blends. Ultrasonic velocity, attenuation and derived acoustic parameters such as adiabatic compressibility, internal pressure and molar sound velocity determine can show finer details of cohesive forces and packing efficiency that would otherwise go unnoticed in conventional macroscopic tests [4]. Pulse-echo and through-transmission ultrasonic techniques have been applied successfully to solutions and solid blends in rubber-based systems to determine compatibility information, phase separation and estimate glass-transition behaviour. Such methods are especially appealing in the case of elastomer blends since the acoustic measurements are directly associated with the elastic modulus and intermolecular forces, and hence can give a mechanistic understanding of blend behaviour [5].

PPG and PEG are polyethers that differ primarily in the molecular weight distribution and the substitution of methyl groups in the backbone; this chemical difference causes distinct hydrogen-bonding and polarity trends, which in turn influence interactions with butyl rubber [6]. Based on composition, molecular weight, and processing history, PPG can produce a range of results, from partial miscibility to distinct phase domains, whereas PEG is highly hydrophilic and forms hydrogen bonds. PPG is less polar and exhibits different solvation behaviour. Earlier ultrasonic and viscometric studies of PPG/PEG systems and polymer-polymer blends have reported the changes in ultrasonic velocity and compressibility with concentration and temperature, and have attributed the changes to miscibility and microstructural reorganization [7]. The available literature provides both empirical and theoretical basis of using acoustic techniques to investigate IIR to PPGA polyblends and IIR to PEG polyblends [8].

The research paper seeks to undertake systematic compatibility tests of butyl rubber/polyether polyblends by the ultrasonic method, complemented by density and thermomechanical factors where applicable. Specific objectives include: (i) measuring ultrasonic velocities and attenuations across a spectrum of frequencies and compositions; (ii) figuring out acoustical parameters that show free volume and intermolecular cohesion; and (iii) comparing acoustic results with anticipated morphological outcomes (compatibility, partial miscibility, or phase separation). Through the convergent consideration of PPG and PEG as blending partners, this piece of work aims at shedding light on the influence of subtle chemical differences between polyethers on interfacial behaviour in non-polar elastomer and the ability of ultrasonic techniques as a diagnostic aid in the development of elastomer blends. The methodological approach maintains the scientific rigor and applicability to real-world formulation issues by drawing on recent characterizations of acoustic polyether and the current ultrasonic compatibility investigation in rubber systems [9].

2. Experimental

2.1 Chemicals used

High molecular viscosity commercial butyl rubber

(IIR/BR) and the analytical-grade polypropylene glycol (PPG) and polyethylene glycol (PEG) were used without further purification. All experiments were done with pure samples of butyl rubber (Polysar, USA) and polyethers having an average molecular weight of 2000 procured as a gift in the Biophysics Laboratory, Central Leather Research Institute (CLRI), Chennai. Liquid blends were made by dissolving the exact amounts of BR in anhydrous toluene to form homogeneous mixtures of stock solution; thereby, 100 : 0, 80 : 20, 60 : 40, 40 : 60, 20 : 80 and 0 : 100 was attained. Polymeric blends were made using solutions 0.5 and 1.0% in benzene, and the mixtures were left to mix over several hours at ambient temperature so as to have complete dispersion of the molecules and so that the air that might have gotten trapped in the mixture could be removed.

All liquid mixtures were identified by their densities, based on the method of gravity (specific-gravity bottle), at 303 K. The relative viscosities of the blend solutions were measured using an Ubbelohde suspended level viscometer held in a constant-temperature bath at 303 K. Measurements of ultrasonic velocity and attenuation were made using a multi-frequency ultrasonic interferometer. A circulating water bath was used to maintain the required isothermal conditions of the sample cell with the blend solution under acoustic measurements at a constant temperature of 303 ± 0.1 K. Each of the compositions was read a number of times to guarantee a reproducible reading. Using the measured ultrasonic velocity (u) and density (ρ), acoustical parameters including adiabatic compressibility, acoustic impedance, and other interaction parameters were determined using conventional thermodynamic relationships to determine compatibility between BR/PPG and BR/PEG liquid mixtures.

3. Results and Discussion

3.1 Viscosity Study, Ultrasonic Study, and Refractive Index Analysis of Blend Compatibility

The tabulation of the acoustic and interaction parameters in Table 1.1 reveals an evolutionary process systematic, composition-dependent, which illuminates the miscibility properties of the BR/PPG and BR/PEG liquid blends. Both blends at 0.5 % concentration show slight and intermediate indications of the parameters, Δb and μ (somewhat negative to slightly positive) indicating a slight perturbation of bulk cohesion, and indicating near-ideal or loosely interacting mixtures at low loading. Increasing the polymer content to 1% gives positive excursions of both PPG and PEG blends with the greatest magnitudes in BR + PEG (Δb and μ both growing rapidly with polyether content). These increases are linked to higher acoustic impedance and decreased adiabatic compressibility, which promote stronger intermolecular association or the onset of microphase structuring rather than merely dilution effects [10]. Since the coefficient of interaction (α) is low across all samples, cohesive density variations, rather than large-scale refractometric contrast, control the most significant acoustic response. Additionally, small differences in refractive index contribute to the limited mixing of molecules at the optical scale [11]. Overall, the ultrasonic signatures of the non-linear velocity/interaction-parameter tendencies and amplified effects at 1% indicate a more significant perturbation and a less optimal mixing in BR/PEG, compared to BR/PPG.

Table 1.1 Interaction Parameters and Refractive Index

Blends	Concentration of BR (wt%)	Δb	μ	α	Refractive Index
BR+ PPG 0.5%	0.2	-0.2022	-0.2856	0.0077	1.4959
	0.4	-0.1648	-0.2327	0.0066	1.4960
	0.6	-0.0427	-0.0603	0.0043	1.4961
	0.8	0.2514	0.3550	0.0016	1.4962
BR+ PPG 1%	0.2	0.7415	0.7579	0.0196	1.4953
	0.4	1.3704	1.4008	0.0166	1.4956
	0.6	2.9184	2.9831	0.0107	1.4959
	0.8	6.0131	6.1464	0.0061	1.4962
BR+ PEG 0.5%	0.2	0.2512	0.3763	0.0058	1.4957
	0.4	0.1651	0.2473	0.0051	1.4960
	0.6	0.0330	0.0494	0.0040	1.4962
	0.8	0.1815	0.2720	0.0021	1.4967
BR+ PEG 1%	0.2	1.4105	1.5106	0.0165	1.4955
	0.4	1.3139	1.4071	0.0159	1.4957
	0.6	2.5539	2.7352	0.0112	1.4960
	0.8	8.0544	8.6260	0.0025	1.4962

To support these observations, the study of viscosity also complements the ultrasonic analysis. The separation of the measured viscosity by the additivity rule results in the interaction parameter Δb which is further elaborated to permit negative intrinsic-viscosity values. The parameters of BR + PPG (1.0%) and BR + PEG (0.5% and 1.0%) (Table 1.1) are always positive, which is a sign of favourable intermolecular interaction and a less supramolecular freedom of the segments in the blended state [12]. Even though BR+PPG at 0.5% has negative values of Δb and μ, the positive α value, which is more appropriate according to Sun et al. (1992), confirms the existence of net attractive interactions [13]. Since incorporation considers all the contributions of interaction, α has a positive sign throughout compositions, which supports the fact that these systems were miscible. Collectively, the viscosity and ultrasonic results have a consistent correlation that BR/PPG and BR/PEG blends are compatible across the concentration range that is studied.

Figure 1.1 shows the dependence of ultrasonic velocity on BR + PPG poly blends with 0.5% and 1% concentration of the poly blends as a function of the blend composition in 303 K. An overall decreasing tendency in velocity can be seen among all systems when the content of polyether rises, which means that the cohesive density becomes lower and the volume of free mixture is larger [14]. This effect indicates the degradation of the intermolecular packing of a butyl rubber structure when flexible PPG or PEG chains enter the rubber structure. The highest initial velocity and deceleration are displayed by BR + PEG (1%) suggesting a stronger initial molecular interaction and a higher degree of structural relaxation at higher PEG content. The relative reduction in BR + PPG blends is smaller, which suggests that the polymer network is perturbing at a relatively more moderate degree, which is in line with a weaker polarity and lesser capacity of hydrogen bonds of PPG [15]. The steady, monotonic decrease of the velocity in the compositions that lacked any sharp deviations, can be interpreted as the lack of the phase separation and indicates the miscible nature of both BR/PPG and BR/PEG systems under the conditions of the study [16-17].

3.3 Adiabatic Compressibility Analysis

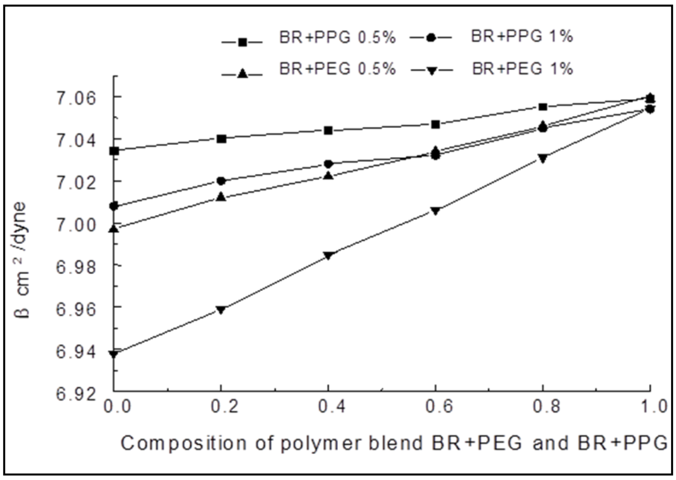

Fig. 1.1 Variation of Adiabatic Compressability with blends BR + PPG and BR + PEG at concentrations 0.5 % and 1% at 303 K.

Figure 1.2 shows the change in the adiabatic compressibility (β) of BR + PPG and BR + PEG blends at the 0.5% and 1% concentration at 303 K. An incremental growth of β is noted with all systems as polyether concentration is increased, which signifies a decrease in cohesive forces and a respective growth in the free volume of the mixtures [18]. This is the same trend directly related to the reduction in the ultrasonic velocity noted above, where the rise in compressibility is generally a sign of more compact, low-density packed molecular environments of the blends, BR + PEG (1%) exhibits the most pronounced effect on increasing β, indicating increased destabilization of the butyl rubber network by the increased polarity and hydrogen-bonding capacity of PEG [19-20]. On the other hand, the comparatively lesser increase in β of BR + PPG blends indicates weaker interaction and more moderate change in microscopic elasticity [21-22]. The gradual and linear change in β with no abrupt changes also indicates the lack of phase segregation and suggests the overall miscibility of BR/PPG and BR/PEG mixtures under the experimental conditions and correlates with the trends expected in ultrasonic compatibility studies of polymer-polymer mixtures.

3.4 Free Volume Analysis

Figure 1. 2 represents the change of the free volume (Vf) of BR + PPG and BR + PEG blends at 0.5% and 1% concentrations with respect to the blend composition at 303 K. There is a steady reduction in the Vf as the ratio of the polyether component increases, suggesting that the packing ability is greater and the molecular mobility of the blend matrix is lower [3, 23].

In BR + PEG systems, this decrease is even more pronounced, particularly at 1% concentration, suggesting the presence of stronger intermolecular interactions because PEG has a higher polarity and hydrogen-binding capacity than PPG [24]. The smoother, phase-uniform Vf reduction across all compositions is consistent with the same lack of phase separation and with earlier ultrasonic parameters that demonstrated blend miscibility. While BR + PPG blends show consistent mixing behaviour, the relatively moderate decrease in Vf suggests weaker specific interactions [25-26].

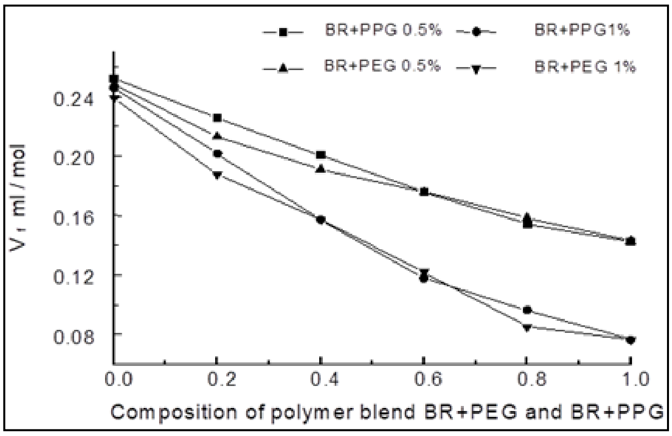

Fig. 1.2 Variation of Free volume with blends BR + PPG and BR + PEG at concentrations of 0.5 % and 1 % at 303 K.

Overall, the trends of free-volume support the evaluation of ultrasonic velocity, compressibility, and viscosity-based tests, all confirming the favorable miscibility of BR with both PPG and PEG at the conditions examined.

3.5 Internal Pressure (Πi) Analysis

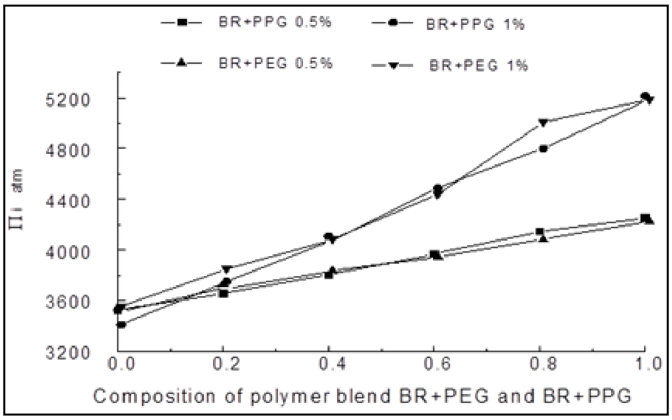

Fig. 1.3 Variation of Internal Pressure with blends BR + PPG and BR + PEG at concentrations 0.5% and 1% at 303 K.

Figure 1.3 depicts the change of the internal pressure (Πi) of BR + PP and BR + PEG blends at 0.5% and 1% concentrations in a series of compositions at 303 K. The overall positive shift in Πi is evident because proportions of PPG and PEG grow, which means that there are more cohesive interactions within the blend matrix. The resultant increase in Πi indicates a loss of free volume and an increase in the number of intermolecular attractions which is corresponding to the literature on the compatibility between polymers and polyethers involving ultrasonic measurements [27-28]. Compared to PPG systems, the Πi of BR + PEG systems shows a much steeper slope, particularly at 1% concentration, suggesting stronger dipolar and hydrogen-bonding interactions that result in a closer molecular packing. Conversely, BR + PPG samples show a moderate, but systematic rise in Πi, thus ruling out a constant miscibility but showing relatively weak specific interactions [29-30]. The continuous transformation of Πi values through all the studied compositions also contributes to the lack of phase segregation. The internal-pressure behaviour, in general, is in good agreement with both compressibility and free-volume outcomes, thus, collectively validating the good compatibility between BR and both PPG and PEG, in that PEG exhibits, comparatively, stronger associative interactions [14, 31-32].

4. CONCLUSIONS

The current analysis indicates the convenience of ultrasonic measurements to determine the compatibility of the butyl rubber (BR) with polypropylene glycol (PPG) and polyethylene glycol (PEG) in blends. Through a methodical assessment of adiabatic compressibility, free volume and internal pressure at temperatures of 303 K with 0.5% and

1% polymer levels; it has been achieved that there are explicit insights on the molecular interactions and miscibility behaviour of the blends. The similarity of the ultrasonic parameters shows that both PPG and PEG react well with BR and no evidence of phase separation is observed in all the compositions studied.

The gradual decrease in the free volume and the increasing internal pressure indicates the creation of smaller sized molecular formations, which indicates the existence of effective intermolecular interactions in the blends. The relative comparison indicates that PEG based blends show a little more interaction with BR as compared to PPG based blends. This discrepancy can be explained by the fact that PEG is more polar and has the ability to form stronger hydrogen-binding and dipolar associations, which results in higher cohesive forces and greater free volume reductions. This interpretation is further supported by compressibility trends which show PEG based systems to exhibit more significant changes in the compressibility coefficient, which is pressure dependent, a measure of heightened structural rigidity and molecular packing as compared to PPG blends. These results confirm known models of polymer-polyether interaction and are also consistent with ultrasonic literature that also observes similar behaviours with compatible elastomer-polymer mixtures.

In general, the ultrasonic method is a sensitive and effective method of investigating the fine details of interaction in BR-polyether systems. The research establishes that BR generates compatible and stable mixtures with both PPG and PEG, and PEG proves to be slightly more compatible. The combined results are not only informative of BR-based polyblends but offer concrete information that leads to the further optimization of elastomeric materials in better performance and, hence, strong material optimization principles may be applied later in future material optimization studies and allow to justify the further applicability of ultrasonic techniques in polymer compatibility research.

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