Evaluation of Mechanical Properties and Physical Interactions of a Ternary Blend of Poly(ethylene-*co*-octene)/Poly(ethylene-*co*-vinyl acetate)/Poly(vinyl chloride) in the Molten State

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ABSTRACT: In this work, ethylene-*co*-vinyl acetate (EVA), poly(ethylene-*co*-octene) (POE), and poly(vinyl chloride) (PVC) blends were processed in a molten state process using a corotating twin-screw extruder to assess both the balance of mechanical properties and physical interactions in the melt state. Tensile measurements, scanning electron microscopy, and oscillatory rheometry were performed. By means of flow curves, the parameters of the power law as well as the distribution of relaxation times were assessed with the aid of a nonlinear regularization method. The mechanical properties for the EVA-POE blend approximated the values for POE, while inclusion of PVC shifted the modulus values to those of neat EVA. The rise in modulus was corroborated by the PVC phase dispersion as solid particles that act as a reinforcement for the ternary blend. The rheological properties in the molten state show that the POE does not present molecular entanglement effects and so tends both to diminish the EVA mechanical properties and increase the fluidity of the blend. However, the addition of PVC both restored the EVA typical pseudoplastic feature and promoted the increase in the viscosity and the mechanical properties of the ternary blend. © 2012 Wiley Periodicals, Inc. J. Appl. Polym. Sci. 129: 1117–1123, 2013

INTRODUCTION

The development of polymeric blends is a practical method which makes that two or more materials of distinct properties attain an intermediary property. In the shoe manufacturing market, this is an usual practice when using elastomers such as poly(ethylene-*co*-vinyl acetate) (EVA) while a second phase, for example, poly(vinyl chloride) (PVC) is incorporated for avoiding wear and improve processability[1]

According to Hernández et al.,[2] numerous works on blends of, for example, PVC/polyolefins and PVC/elastomers have been published. The focus on PVC blends is due to the fact that PVC is a very versatile polymer, useful in several domains such as medicine, civil construction, clothing, fashion, and packaging.

The EVA copolymer belongs to a statistical copolymers class having intermediary properties between its two constituting homopolymers, polyethylene and poly(vinyl acetate) (PVAc). Such intermediary properties derive from its complex morphology, which comprises a crystalline phase (with ethylene units), an interface region (with ethylene moieties and VAc moieties), and a complex amorphous phase (with ethylene moieties and noncrystallized VAc units).[3] As the EVA crystallinity is a function of the VAc content, the stiffness of the EVA copolymer will depend on the size of the ethylene crystallizable phase. Poly(ethylene-*co*-octene) (PEO) is an ethylene-based copolymer having excellent compatibility with thermoplastic olefins. Its elastomeric features derive from the octene comonomer. The crystallinity reduction and augmented copolymer flexibility[4] resulting from the hexane group branching enable PEO to be used in blends to improve the impact strength and the low temperature brittleness.

In blends of polymers of different chemical character such as EVA, PVC, and POE, several criteria should be taken into consideration. The basic criteria related to the miscibility and compatibility between polymers is dependent on the particular structure of each material. Also, due to the nature of the long

macromolecular chains, the dynamic interdiffusion of the molecules is considerably slow.[5]

PVC and EVA interact satisfactorily for contents above 45% VAc[6,7] alone; on the other hand, using high contents of VAc in a blend with PVC can hinder the formation of ethylene moieties crystals. So, aiming at a ternary blend, the use of POE could toughen low content-VAc EVA while PVC would bring improved processing by acting as reinforcement resulting in improved mechanical properties. The aim of this work is to study the mechanical and rheological properties in the melt state resulting from the incorporation of PVC in a blend of EVA and PEO.

MATERIALS

The materials used in this study were poly(ethylene-co-vinyl acetate) Evateno® HM 728 containing 28% of vinyl acetate (EVA-28), supplied by Braskem, Brazil (MFI: 6.0 g/10 min according to ASTM D 1238); PVC Norvic® SP1000, supplied by Braskem Brazil (K value 65 ± 1, suspension polymerization resin, with high porosity and narrow granulometric distribution); PVC (MFI = 0.33 g/10 min) Norvic® SP1000, supplied by Braskem Brazil (K value 65 ± 1, suspension polymerization resin, with high porosity and narrow granulometric distribution); and the elastomeric copolymer ethylene-co-octene (POE) (Dow Engage® 8130; MFI = 3.87 g/10 min) obtained from The Dow Chemical Company. The molten point of the polymers is 160, 124, and ≈180°C for EVA, POE, and PVC, respectively.

METHODS

Sample Preparation

Blends were prepared in weight percent (wt %). The blends were prepared with 50/50 wt % of EVA/POE and replaced 5, 10, 15, and 20 wt % of POE by PVC. After drying the samples in an oven with air circulation at 60°C for 8 h, the blends were processed in a molten mixture process using a corotating twin-screw extruder (MH-COR-20-32-LAB, MH Equipments; $D = 20$ mm, L/D = 32) with eight heating zones and temperature profiles of 100, 120, 140, 160, 130*, 160, 160, and 160°C (barrel to matrix, respectively) and a processing speed of 400 rpm. In zone 5* vacuum degassing was used.

Mechanical Properties

The tensile strength tests were performed on type IV specimens (ASTM D 638), using an extensometer with a maximum elongation of 250 mm and a 2 kN load cell (EMIC DL 3000).

Oscillatory Rheometry

The samples were analyzed in an Anton Paar oscillatory rheometer (Physica MCR 101). The plate–plate geometry with 25 mm diameter and 1 mm of distance between the plates was used. The analysis was carried out at a frequency range of 0.1–100 Hz, shear stress maximum of 200 Pa, and nitrogen flow of 1 m^3/h. The temperatures used to analyze the pure polymers were 160, 125, and 180°C for EVA, POE, and PVC. For all blends, 160°C was used. From the results of the rheological analysis, the Cox-Merz relationship was calculated,[8] and relaxation ($H(\tau)$) spectra were obtained using the nonlinear regularization (NLREG) program.[9–12]

Viscoelastic Functions

In the molten state, two types of behavior can be considered. The linear and nonlinear viscoelasticity directly affect the response to deformation of the materials. At low amplitudes of deformation, under linear viscoelastic regimen, the relaxation modulus can be obtained for all levels of deformation.[13] However, in the study of viscoelasticity, several physical functions, such as relaxation $H(\tau)$, cannot be directly obtained from experiments.[14] Nevertheless, in many cases, these can be obtained from an experimentally measurable quantity through the resolution of an inverse problem. Through the use of the Tikhonov regularization in the nonlinear generalization for inverse problems, called the nonlinear regularization method and used in the NLREG program, it is possible to obtain functions such as the relaxation ($H(\tau)$) and retardation ($L(\tau)$) spectra.[9,10] These viscoelastic functions can be obtained through storage modulus (G') and loss modulus (G'') measurements using the relations described in eqs. (1) and (2)[11,12]:

$$G'(\omega) = G_\theta + \int_{-\infty}^{\infty} H(\tau) \frac{\omega^2 \tau^2}{1 + \omega^2 \tau^2} d \ln \tau \tag{1}$$

$$G''(\omega) = \int_{-\infty}^{\infty} H(\tau) \frac{\omega^2 \tau^2}{1 + \omega^2 \tau^2} d \ln \tau \tag{2}$$

where: G_θ is the equilibrium modulus, $H(\tau)$ the relaxation spectrum function, and τ is the relaxation or retardation time.

The relaxation spectrum is the result of Maxwell's sum of infinite elements which describes the elastic behavior of the polymer, as well as its ability to store energy. The estimation of the retardation spectra is carried out through inter-relations of the spectra described by Ferry,[13] which depict the viscous (imaginary) portion of the system, as represented in eqs. (3) and (4), respectively.

$$H(\tau) = \frac{L(\tau)}{\left[J_g + \int_{-\infty}^{\infty} \frac{L(\tau)}{(1-\tau)/\tau} d \ln \tau - \frac{\tau}{\eta_0} \right]^2 + \pi^2 L(\tau)^2} \tag{3}$$

$$L(\tau) = \frac{H(\tau)}{\left[G_\theta - \int_{-\infty}^{\infty} \frac{H(\tau)}{\tau/(\tau-1)} d \ln \tau \right]^2 + \pi^2 H(\tau)^2} \tag{4}$$

where J_g is the instantaneous compliance and η_0 the zero shear-rate viscosity.

Scanning Electron Microscopy

Scanning electron microscopy (SEM) was carried out using a Superscan S-550 microscope, with a secondary electron detector and an acceleration voltage of 15.0 kW. The scanning was carried out at a magnification of 3000× (scale of 5 μm). The samples were previously covered with gold.

RESULTS AND DISCUSSION

Figure 1 shows the tensile test results of pure polymers and their blends. Young Modulus is higher for EVA relative to POE and lower than those of PVC. Stress values follow the same pattern as the Modulus. PVC has the lowest elongation values among the three polymers and POE values are higher than those of EVA. In the EVA/POE (50/50/0) wt % blend, the mechanical properties values are close to those of neat POE. The trend resulting from variation in composition of the EVA and
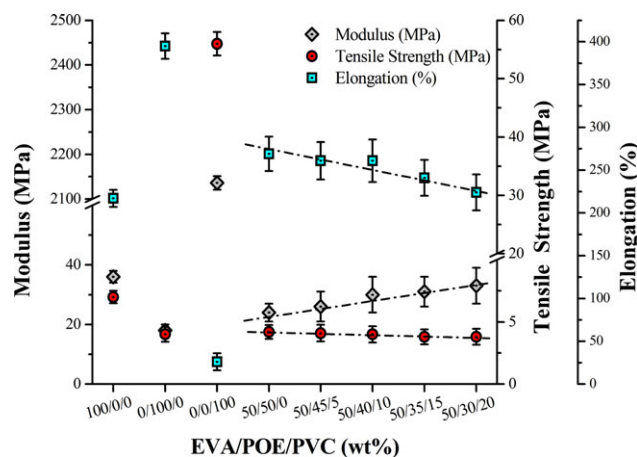
Figure 1. Mechanical properties values obtained by tensile test. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

POE blend is a function of the amount of POE incorporated into EVA. The replacement of 5, 10, 15, and 20 wt % of POE by PVC has approximated the modulus values to those of neat EVA as PVC is a rigid polymer, which in this case was mixed below its molten point, ≈180°C. The same behavior was observed for the elongation values while the stress values were kept constant. The mechanical properties measurements for the ternary blend show that the incorporation of PVC tends to compensate the loss in toughness by the flexibility of POE blended to EVA.

The morphology of the pure EVA/POE and the EVA/POE/PVC blends were showed in the Figure 2. The Figure 2(a) showed homogeneity between the EVA and POE due to the interaction of the ethylene phase of EVA with the POE. As seen in the (b), (c), (d), and (e) images in Figure 2, PVC is a separate phase, which works as a reinforcement as the PVC particles are dispersed below the molten point. The shifts in properties of the EVA/POE blend toward of the neat EVA resulting from the POE replaced by PVC. Besides also may be associated with particle dispersion and homogeneity of the PVC fraction incorporated.

If one assumes homogeneity between EVA and POE and the PVC incorporation represents a reinforcement phase, then the ternary blend is able to assure both easier processing by incorporating POE into EVA and the balance of properties seen in Figure 1 resulting from the PVC incorporation.

The POE addition is a strategy to increase the fluidity and facilitate the processing of the EVA. For consequence, the POE promotes a higher flexibility in the mixture. A fixed proportion of 50/50 wt % of EVA/POE was used due to the fact that concentration has retained the best balance between the mechanical properties (the results of the variation of the concentration between the EVA and POE are not presented in this study). The 50/50 wt % of EVA/POE was used as starting point for evaluating the influence of PVC as a third phase in the mixture.
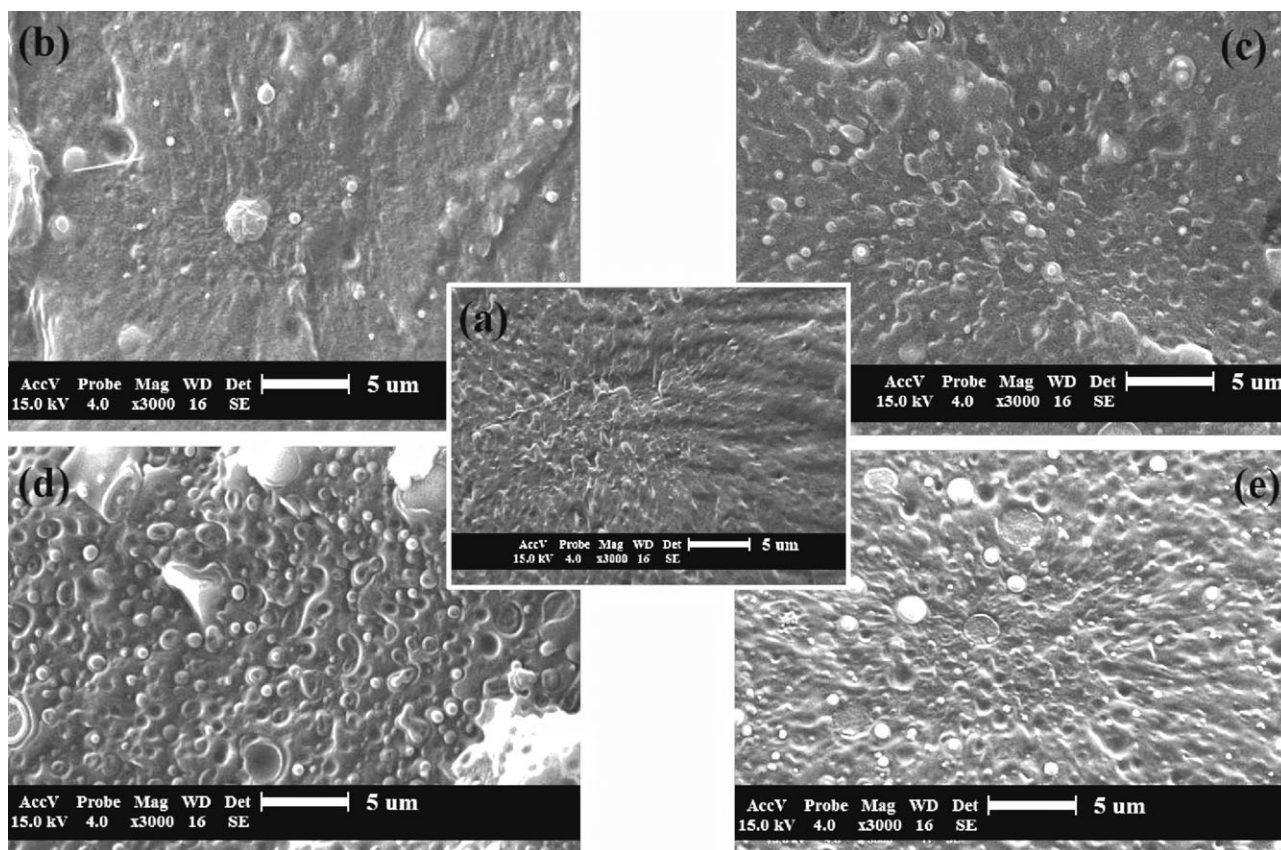

Figure 2. SEM analysis obtained for the EVA/POE and EVA/POE/PVC blends were: (a) 50/50/0, (b) 50/45/5, (c) 50/40/10, (d) 50/35/15 e (e) 50/30/20.
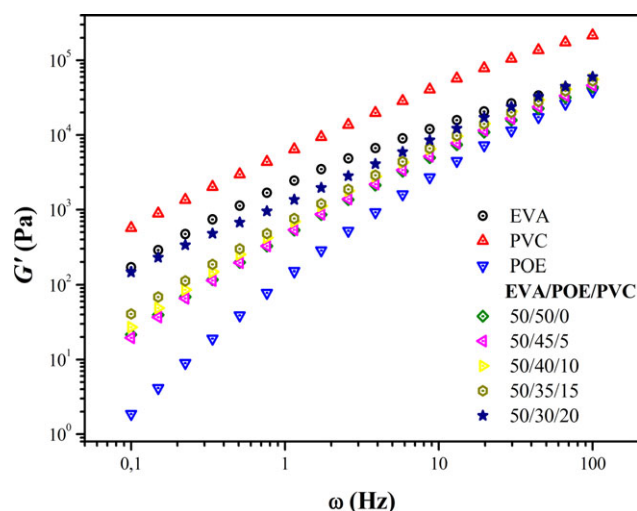
Figure 3. Storage modulus for the pure components and their blends. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

To assess the behavior of these blends in the melt state and understand the physical interaction effects between the phases, Figure 3 depicts the values for the storage modulus (G') while Figure 4(a,b) illustrate modulus values recorded for the frequencies of 0.1, 1, 10, and 100 Hz, respectively. It was observed that PVC has the highest values for G' except for the frequency 1.0 Hz. EVA values are lower than those for PVC, but higher than those for POE, mainly at low frequencies.
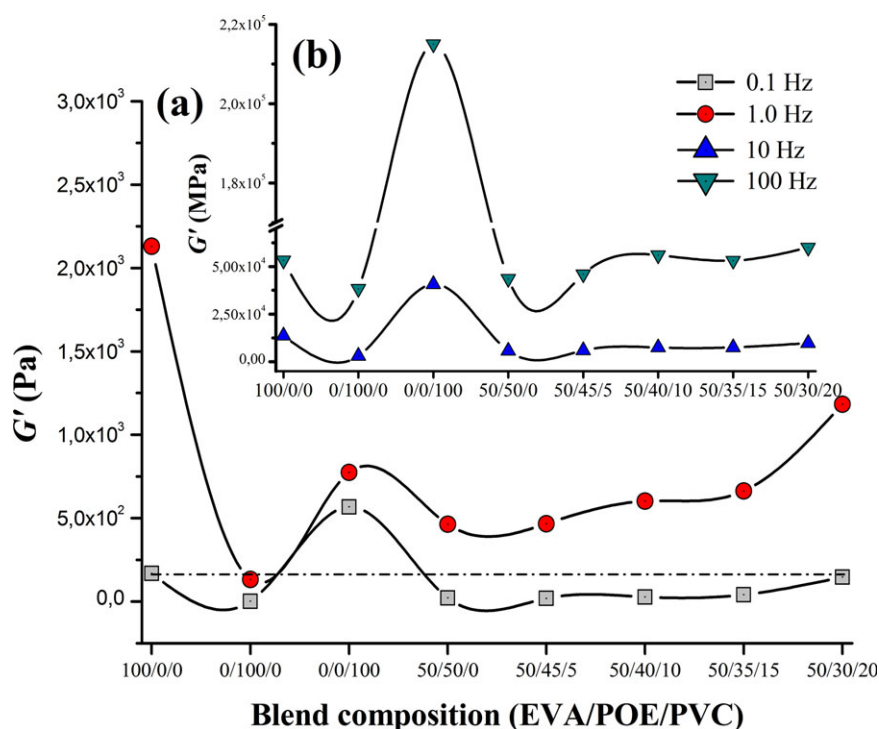
The modulus values of EVA/POE blend with 50 wt % of EVA content were close to those of POE chiefly at low frequencies. Yang et al.[15] studied blends of thermoplastic polyurethane (TPU) and POE and reported that the interface between the phases of the blend may be influenced by the TPU hydrogen bonds. As POE is able to increase the free volume between the chains or reduce the quantity of interaction points (associated with the reduction of the viscosity), this material entails weak secondary intermolecular forces and thus a weak interface in the blend with EVA is to be expected, leading to a reduction in G'.

PVC is a rigid polymer. Such rigidity is associated with the electronegativity of the chlorine atoms that hinder its molecular conformation. The addition of PVC in the EVA/POE blend showed a trend toward increased G'. The addition of 20 wt % of PVC led to values near the EVA. This suggests that PVC is able to compensate the flow stability related to the energy loss when it is added to the EVA/POE blend. The flow stability can be interpreted through the kinematic viscosity curves obtained by the Cox-Merz relationship as depicted in Figure 5.

The flow curves provide a better understanding of the phenomena resulting from processing, as viscosity is found to be a function of the shear rate ($\dot{\gamma}$). Analysis of the viscosity curves as a function of the shear rate reveals a tendency similar to that of the curves in Figure 3.

However, it is possible to add that the viscosity tends to approach the values of POE for the 50/50/0 wt % blend. The presence of diluents and plasticizers in polymer systems can increase the adhesion in the stationary state and reduce the viscosity when the shear rate is equal to zero.[8] In this case, it is

Figure 4. Points of storage at different frequencies for the pure components and their blends. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]
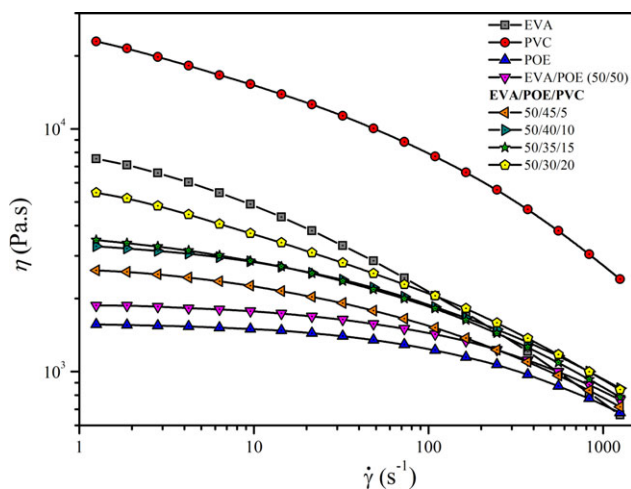
Figure 5. Kinematic viscosity vs. shear rate curves obtained from the Cox-Merz relationship. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

Table I. Power Law Parameters for Homopolymers and Blends

EVA/POE/PVC (wt %)	n	K	r
100/0/0	0.64	50485	0.9970
0/100/0	0.89	23472	0.9986
0/0/100	0.69	80146	0.9965
50/50/0	0.88	25477	0.9986
50/45/5	0.82	30157	0.9983
50/40/10	0.81	33503	0.9981
50/35/15	0.79	34425	0.9980
50/30/20	0.73	41428	0.9989

probable that the role of POE in the blend is as a diluent or plasticizer.

Both for low and high shear rates, the PVC addition raised the viscosity values in the blends. This rise in flow stability suggests that PVC can act to limit the flow of EVA and POE chains. Lizymol et al.[16] state that it is already known that the EVA/PVC blends are miscible in systems in solution. This observation can be taken into consideration for the samples in the melt state, as the higher degree of freedom in the melt state can favor the interaction between VAc groups and PVC. The flow curves (Figure 5) enables the application of the power law[8] as described in eq. (5).

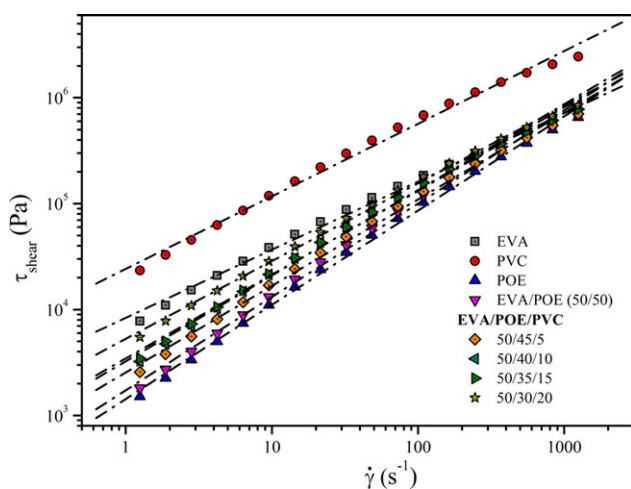$$\tau_{\text{shear}} = K\dot{\gamma}^{n} \quad (5)$$

Figure 6. Linear fit for determination of power law parameters. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

where τ_{shear} is the shear stress, K is the consistency index, that is, it is the viscosity value when the shear rate is equal to zero, $\dot{\gamma}$ is the shear rate and n is the power law index. The power law index (n) is a measure of the pseudoplasticity of polymers and is thus a consequence of the entanglement effect, molecular orientation, and also the generation of heat by shearing. From an analysis of n, it can be observed that the closer the value of n is to zero, the more pseudoplastic is the material.[8] By applying eq. (5), Figure 6 shows the fits acquired to determine the parameters of the power law.

The fits of Figure 6 give a correlation coefficient (r) close to unity, i.e., $r \approx 1$, demonstrating a good linear relation. The minimum and maximum values of r obtained at a confidence interval of 95% were $r = 0.9978$ and 0.9997, respectively. The results attained are given in Table 1.

Among the polymers studied, EVA has the highest and POE the lowest pseudoplasticity character. Pseudoplasticity is associated with the degree of physical entanglements between chains. Therefore, one can assume that the rate of EVA viscosity loss is more pronounced as a result of the presence of more entanglement effects between chains. POE does not undergo entanglement probably because of space impeachment between chains caused by the size of the octene branching. In this case, POE approaches the behavior of a Newtonian fluid.

The index of consistency is higher for EVA than for POE and this means that the higher viscosity reflects higher molecular entanglement as already mentioned. The n value for PVC is intermediary to those for EVA and POE, however, the value for K is higher than those for EVA and POE. The higher PVC stationary viscosity results from the monomer units conformation obstacle caused by the high PVC polarity that hinders the mobility of molecule segments.

The n and K values for the 50/50/0 wt % blend were close to those of POE as seen in the already discussed results. These results suggest that the predominance of the Newtonian fluid behavior is due chiefly to the amount of POE and less to the EVA entanglement effects. In this case, even if one cannot distinguish two phases in the SEM image of Figure 2(a), it is hypothesized that there are restrictions or hindrances for EVA to interact with POE in the melt state. This immiscible phase in the melt state suggests a lower critical solution temperature (LCST) behavior. In general, polymer–polymer blends have a
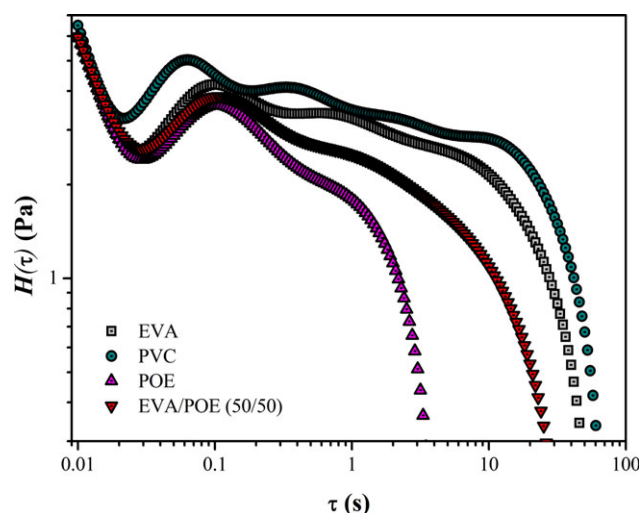
Figure 7. Relaxation spectra illustrating the distribution of the relaxation times for pure components and EVA/POE blend. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

LCST behavior caused by diffusional problems resulting from the molecular rearrangement being hindered by the extension and size of the chains; that is why at low temperatures the blend can be soluble.

To understand the effects of the interaction between the phases of the blend, the study of the viscoelastic functions such as the relaxation $H(\tau)$ and retardation $L(\tau)$ spectra allows a more detailed analysis of the flow phenomena in the melt state.

It is well-known that relaxation phenomena are strongly dependent on both the molecular weight and the architecture (branching) of the system.[17] It is known that the chain segments tend to relax by cooperativity.[13] Cooperativity, in turn, involves rearrangements of several segments of the main chains over time. Over very long time periods, considering the melt state, disarticulation of the chains occurs through rupture of topological effects such as chain entanglements.[18] Thus, by analyzing a relaxation spectrum relating to the melt state, it is possible to evaluate the effects related to the flow behavior of some material.[11] As viscosity is one of the main characteristics of polymeric materials, it is important to evaluate the effects of relaxation time distribution, $H(\tau)$.[18] Figures 7 and 8 show the relaxation ($H(\tau)$) spectra obtained for the samples studied.

Based on the relaxation curves obtained (Figure 7), there are three main relaxation phenomena in the melt state. According to Matsuoka,[18] the first stage is related to the conformational changes, that is, the melt state is characterized by the state of highest entropy where the chains have a sufficient degree of freedom to vibrate and undergo conformation along the structure. At this stage, it is important to consider that the type of binding atom, particularly in the main chain, can alter the conformation effects due to differences in electronegativity and thus the mobility of the main chain. The second stage is related to chain sliding. This phenomenon is related to the type of structure, for example, the quantity of branches and intermolecular forces.

According to Privalko et al.,[19] in terms of the reptation model of polymer flow, the variation in $H(\tau)$ observed in the second stage of relaxation may be associated with the complete escape of a polymer chain from the "virtual" tube formed by the neighboring chains. Finally, the third stage of relaxation, according to Matsuoka,[18] is associated with the sliding of the chain entanglements, given the inhibited passage or escape of the chains from the "virtual" tube described by the reptation model.

In Figure 7, the neat EVA portrays the three molecular relaxation phenomena at approximately 0.1 s (molecular conformation), 1 s (chain sliding), and 10 s (entanglement movement). POE showed only the first two steps at approximately 0.1 and 1 s. The absence of chain entanglement sliding corroborates both the behavior close to that of a Newtonian fluid and the observation that the space impeachment between chains makes impossible physical entanglements between copolymer chains to occur. The PVC relaxation peak was at approximately 0.04 s followed by other three peaks at 0.3 s, 2 s, and 10 s. PVC molten is not homogeneous as PVC has a particle structure that hinders the occurrence of the complete molten process.[20] This is consequent to a particle structure formed during polymerization, that is, the polymer is not soluble in its own vinyl chloride monomer. This feature leads to the formation of a nonhomogenous molten mass with particles trapped between the molten polymer. The first two relaxation peaks at 0.04 s and 0.3 s can be associated with the phases of molten and solid PVC particles. This metastable feature could explain the separated relaxation and conformation times between the PVC solid/liquid phases. The further relaxation peaks, 2 s and 10 s are associated with the same relaxation phenomena observed for EVA. For PVC instead, the relaxation phenomena are shifted to longer times. In PVC, the intensity of the peak associated with the chain sliding is lower than that of EVA; however, the relaxation peak associated with the displacement of entanglements is clearer.
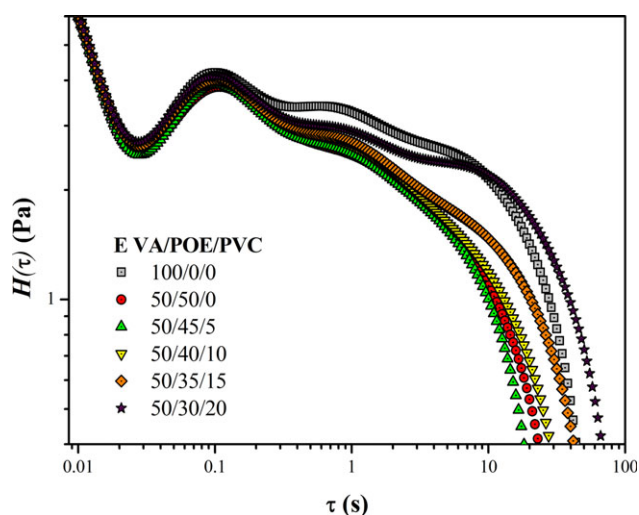

Figure 8. Relaxation spectra illustrating the distribution of the relaxation times for the blends studied. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

As PVC bears a huge concentration of bulky chlorine atoms that increase chain rigidity, chain sliding is hindered and the displacement of entanglements outweighs that of individual chains.

The relaxation times of the binary blend of EVA and POE were similar to those of neat POE; however, the distribution of the chain sliding phenomena is displaced toward longer times. This particular binary blend corroborates the fact that POE restricts the effects of physical entanglement and thus makes the chain flow easier. Upon incorporating PVC, again the relaxation phenomena linked to entanglement sliding could be observed, see Figure 8.

For the EVA/POE/PVC blend containing 50, 30, and 20 wt % of EVA, PEO, and PVC contents, respectively, this third relaxation phenomenon appeared even more pronounced than for neat EVA. The appearance of this entanglement sliding phenomenon for the ternary blend shows that the PVC restoring the mechanical properties and introducing once more and interaction interface with the EVA/POE blend. The separated phase of the PVC performs a counterbalance with the ability of the POE increasing the fluidity by reduction in the physical entanglements effects in the EVA-containing blend. Such features may be responsible for the improved flow stability and maintenance of the desired mechanical properties such as the low content-VAc EVA toughness, POE impact strength, and reinforcement of the elastic modulus and thermal stability resulting from the PVC phase.

CONCLUSIONS

Tensile measurements evidenced that the 50/50 binary blend of EVA and POE produced mechanical property data close to those of neat POE; however, the elongation values were intermediary to the two phases. Replacement of POE by PVC indicates a trend in rise of the elastic modulus because the PVC is a rigid particle incorporated into the solid state and causing the reinforcing effect in the EVA/POE blend. The observed modulus rise led to values that are close to those of neat EVA.

The reinforcement effect was ascertained by the dispersion of the PVC phase in the SEM images. Despite the fact that no phase differences between EVA and POE could be determined, PVC formed a separate phase due to the characteristic rigid particles and it be incorporated at a temperature lower than its molten state.

Oscillatory rheometry tests indicate that the values of G' and η showed the same dependence observed by the elastic modulus on mechanical testing. The EVA/POE blend evidenced that the feature of POE of being devoid of molecular entanglement effects leads the blend of these two phases to a behavior close to that of a Newtonian fluid, characteristic of neat POE. Therefore, the EVA/POE blend manifests that these polymers are not miscible in the melt state. By incorporating PVC into the binary blend, the behavior observed for neat EVA is approached. In the ternary blend compositions (from 50/50/0 to 50/30/20), the n value decreases by the incorporation of PVC (of lower n value) or by decreasing the POE content (of higher n value) or by a combination of both.

Molecular relaxation spectra data support the rheological results demonstrating that the relaxation times of the EVA/POE/PVC ternary blend assure the balance between the polymers characteristics. In other words, the POE promotes a reduction of the viscosity in the blend with the EVA, however, the mechanical properties are close the POE, on the other hand, the inclusion of PVC as a third phase showed an reinforcement effect between the EVA and the POE.

REFERENCES

1. Mcconnell, D. C.; Mcnally, G. M.; Murphy, W. R. *J. Vinyl Addit. Technol.* **2002**, *8*, 194.

2. Hernández, R.; Peña, J. J.; Irusta, L.; Santamaría, A. *Eur. Polym. J.* **2000**, *36*, 1011.

3. Yamaki, S. B.; Prado, E. A.; Atvars, T. D. Z. *Eur. Polym. J.* **2002**, *38*, 1811.

4. Lee, S.-H. *J. Appl. Polym. Sci.* **2005**, *95*, 161.

5. Datta, S.; Lohse, D. J.; Polymer Compatibilizers Uses and benefits in Polymer Blends; Hanser Publishers: New York, **1996**.

6. Righetti, M. C.; Cardelli, C.; Scalari, M.; Tombari, E.; Conti, G. *Polymer* **2002**, *43*, 5035.

7. Bureau, E.; Cabot, C.; Marais, S.; Saiter, J. M. *Eur. Polym. J.* **2005**, *41*, 1152.

8. Riande, E.; Díaz-calleja, R.; Prolongo, M. G.; Masegosa, R. M.; Salom, C. Polymer Viscoelasticity: Stress and Strain in Practice; Marcel Dekker: New York, **2000**.

9. Roths, T.; Marth, M.; Weese, J.; Honerkamp J. *J. Comput. Phys. Commun.* **2001**, *139*, 279.

10. Weese, J. *Comput. Phys. Commun.* **1993**, *77*, 429.

11. Pistor, V.; Lizot, A.; Fiorio, R.; Zattera, A. *J. Polymer* **2010**, *51*, 5165.

12. Pistor, V.; Ornaghi, H. L., Jr.; Ferreira, C. A.; Zattera, A. J. *J. Appl. Polym. Sci.* **2011**,*125*, 462.

13. Ferry, J. D. Viscoelastic Properties of Polymers; Wiley: New York, **1980**.

14. Pistor, V.; Chiesa, A.; Ornaghi, H. L.; Fiorio, R.; Zattera, A. *J. Int. Polym. Process.* **2011**, *1*, 383.

15. Yang, J.; Chen, X.; Fu, R.; Zhang, M.; Chen, H.; Wang, J. *J. Appl. Polym. Sci.* **2008**, *109*, 3452.

16. Lizymol, P. P.; Thomas, S.; Jayabalan, M. *Polym. Int.* **1997**, *44*, 23.

17. Brabec, C. J.; Schausberger, A. *Rheol. Acta* **1995**, *34*, 397.

18. Matsuoka, S. Relaxation Phenomena in Polymers; Hanser Publishers c: Munich, **1992**.

19. Privalko, V. P.; Shumsky, V. F.; Privalko, E. G.; Karaman, V. M.; Walter, R.; Friedrich, K.; Zhang, M. Q.; Rong, M. Z. *J. Mater. Process. Technol.* **2003**, *137*, 208.

20. Wilkes, C. E.; Summers, J. W.; Daniels, C. A. PVC Handbook; Carl Hanser Verlag: Munich, **2005**.