Physical Properties of Ethylene Vinyl Acetate Copolymer (EVA)/Natural Rubber (NR) Blend Based Foam

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ABSTRACT: In this study an attempt was made to improve the rebound resilience and to decrease the density of ethylene-vinyl acetate copolymer (EVA) foam. For this purpose, EVA was blended with natural rubber (NR), and EVA/NR blends were foamed at 155°C, 160°C, and 165°C. To investigate the correlation between crosslinking behavior and physical properties of foams, crosslinking behavior of EVA/NR blends was monitored. The physical properties of the foams were then measured as a function of foaming temperatures and blend compositions: 165°C was found to be the optimal temperature for a crosslinking of EVA/NR foam. As a result, the density of EVA/NR blend foamed at 165°C was found to be the lowest. EVA/NR (90/10) blend, foamed at 165°C, showed lower density, better rebound resilience, and greater tear strength than EVA foam. © 2004 Wiley Periodicals, Inc. J Appl Polym Sci 94: 2212–2216, 2004

Key words: foam; EVA; NR; crosslinking, density

INTRODUCTION

Because of advantages of light weight, buoyancy, cushioning performance, thermal and acoustic insulation, impact damping, and cost reduction, the markets for foams have been growing rapidly worldwide. To develop foams with improved properties and lower cost, it is important to understand the relationship between parameters of foam formation and physical properties of the foams.[1]

Nowadays ethylene-vinyl acetate copolymer (EVA) foam is widely used for many purposes (e.g., in the midsole, the layer that lies between insole and outsole of running shoes).[2] In this study, our focus was to improve the rebound resilience and decrease the density of EVA foam; and for this reason, EVA was blended with natural rubber (NR).

To obtain optimal foam expansion and good physical properties of the foams, optimal crosslinking is the most critical requirement.[3–6] In this study, crosslinking behavior of the EVA/NR blends was observed at various temperatures (155°C, 160°C, and 165°C) using an oscillating disk rheometer (ODR). To investigate the relationship between crosslinking behavior and physical properties of foams, foams were prepared at 155°C, 160°C, and 165°C, based on EVA/NR blends, and the physical properties of the foams were measured as a function of foaming temperatures and natural rubber compositions.

The detailed knowledge of the effect of composition and foaming parameters on the physical properties of the foam is important to control and optimize the physical properties of EVA/NR foam. The effects of foaming temperatures and blend compositions on density, rebound resilience, tensile strength, tear strength, and cell structure of the foam were investigated in the present study.

EXPERIMENTAL

Materials and foam preparation

Names and important characteristics of the materials used in this study are summarized in Table I. Figure 1 shows the schematic diagram of the experimental procedure. EVA, NR, and other additives were mixed in a Haake internal mixer at 140°C for 15 min. EVA was first allowed to melt, followed by the addition of NR and other additives. Then the obtained EVA/NR blends were mixed with chemical blowing agent and crosslinking agent in a two-roll mill. DCP (crosslinking agent) content was fixed at 0.6 phr based on the total amount of EVA and NR. The blowing agent used was azodicarbonamide-based blowing gas release system. Azodicarbonamide is odorless and easily dispersed. It is activated by organic acids, bases, and metal compounds.

After mixing in a two-roll mill, the mixture was put in a mold and the foams were obtained by compression-molding. To investigate the effect of foaming

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TABLE I
Characteristics of Materials Used in this Study

Materials		Characteristics	Source
Resin	EVA	VA content: 19%, Melt index: 1.8	Hanwha
	NR	SMR L	Astlett Rubber
Crosslinking agent	DCP	Purity: 99%	Akzo Nobel
Blowing agent	JTR/M	Decomposition temperature: 138 ≈ 144°C	Kum Yang
Blowing co-agent	ZnO Stearic/acid		LG Chem.
Filler	$MgCO_3$	Specific gravity: 2.958	Tokuyama

temperature on the physical properties of EVA/NR foam, the foaming was carried out at three different temperatures (155°C, 160°C, and 165°C). The mixture was pressed at 14.7 MPa, in a hydraulic press, at 155°C, 160°C, and 165°C for 40 min, respectively. After removal of the pressure, expansion takes place immediately, and the obtained foams were left at room temperature to cool down.

MEASUREMENTS

Density of the foam was measured by a buoyancy method using a gravimeter (Ueshima, MS-2150). In the measurement, the samples used were 1 × 1 cm^2 of area and the thickness 1 cm. The samples were weighed first in air, and then in water. The density of the foam was determined using Archimedes' principle.

Cure characteristics were studied with an Oscillating Disk rheometer (Gotech) according to ASTM D2084. (The rheometer rotor oscillates through 1° arc at 1.66 HZ and exerts a shear strain on the sample during the curing of the sample.)

A Universal Testing Machine (Model 4466, Instron Co.) was used to obtain the tensile strength of the EVA/NR foams at room temperature. The crosshead speed was 500 mm/min. All measurements were performed for five replicates of dog-bone-shaped specimens and averaged to get the final result. Also, the tear strength was measured using unnicked 90° angle test pieces at a crosshead speed of 500 mm/min in the Universal Testing Machine.

The rebound resilience (elasticity) was measured according to DIN 53512. (The pendulum is released from a horizontal position and strikes the samples at a vertical point.) To investigate cellular structure and apparent mean cell diameter, the cross sections of the EVA/NR foams were microtomed at low temperature and were examined with a Scanning Electron Microscope (JEOL JSM-35CF).

RESULTS AND DISCUSSION

The selection of temperature is very important for optimal crosslinking. To select optimal crosslinking temperature, crosslinking behavior was monitored by an oscillating disk rheometer (ODR). The effects of temperatures and blend compositions on the

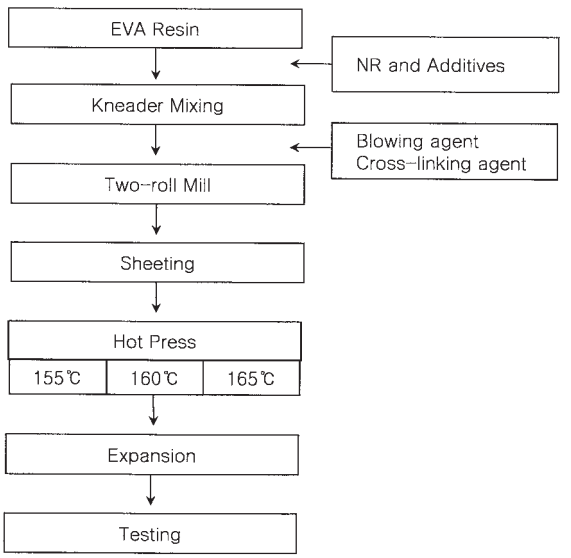

Figure 1 Schematic diagram of EVA/NR foam preparation.

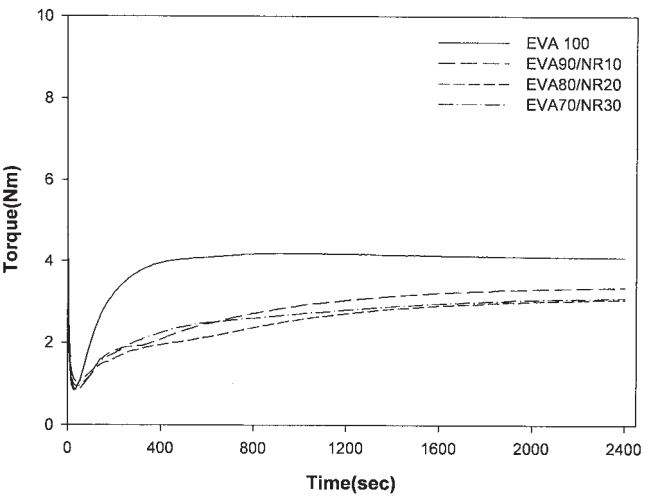

Figure 2 Crosslinking behavior of EVA/NR blend at 155°C.

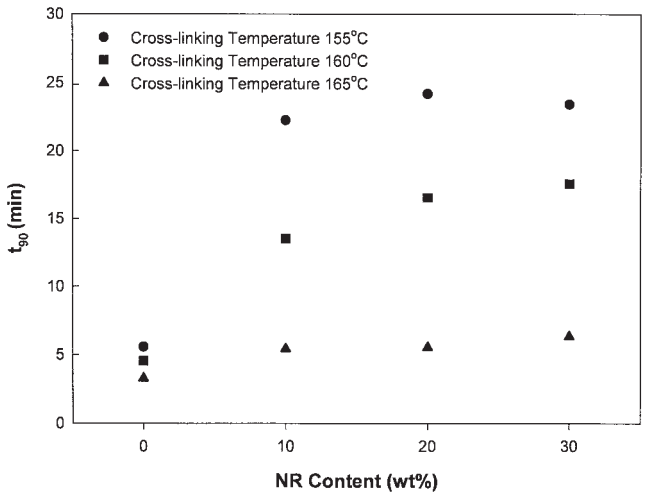

Figure 3 Relationship between t_{90} and blend composition at various temperatures.

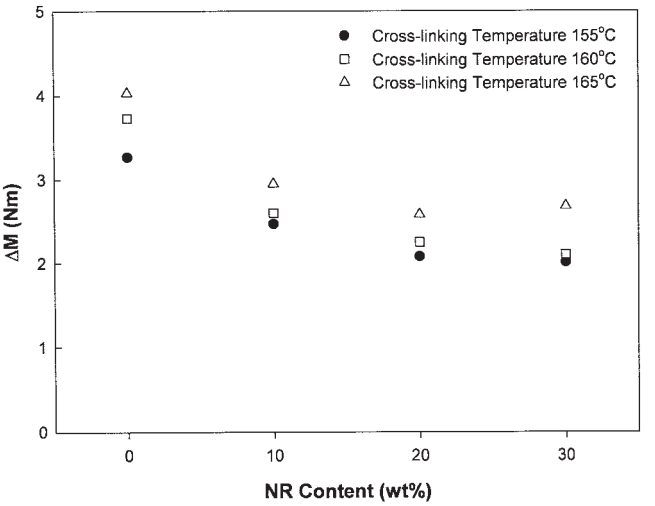

Figure 5 Dependence of torque difference on blend composition and temperatures.

crosslinking characteristics of EVA/NR blend were examined at various temperatures (155°C, 160°C, and 165°C) and various compositions (100/0, 90/10, 80/20, 70/30).

Figure 2 shows the rheographs obtained for various compositions of EVA/NR blend at 155°C as an example. Both the maximum torque and the difference between the maximum and minimum torque decrease with addition of NR into EVA. Since maximum torque generally correlates with hardness and/or modulus, the incorporation of NR decreases the stiffness of the blend. More torque difference indicates more crosslink density.[7] Therefore, the decrease of the torque difference, with addition of NR, indicates lower crosslink density (i.e., the incorporation of NR in the blend decreases the crosslink density).

Figure 3 shows the variation of the cure time, t_{90} (time required to reach 90% crosslinking) of the EVA/NR blends, with composition at various crosslinking temperatures. At 155°C and 160°C, t_{90} increases significantly with increasing NR content up to a certain level in the blend. This indicates a slower crosslinking reaction with NR addition. However, at the same blend ratio, t_{90} of the EVA/NR blend at 165°C is much shorter than that at 155°C and 160°C. The crosslinking reaction of the EVA/NR blend at 165°C is much faster than at 155°C and 160°C.

Figure 4 shows that maximum torque decreases with increasing NR composition and decreasing crosslinking temperature. Since maximum torque generally correlates with hardness and/or modulus, it can be concluded that the incorporation of NR and the reduction of crosslinking temperature lead to a decrease in the stiffness of the blend. The decrease of the stiffness of the blend with reducing crosslinking temperature could be due to the decrease of crosslink density at lower crosslinking temperature.

The difference between the maximum and minimum torque (ΔM) for EVA/NR blends decreases with increasing NR composition and decreasing crosslinking temperature (Fig. 5). As mentioned earlier, the torque difference is a reflection of crosslink density.[7] Therefore, it can be concluded that the incorporation

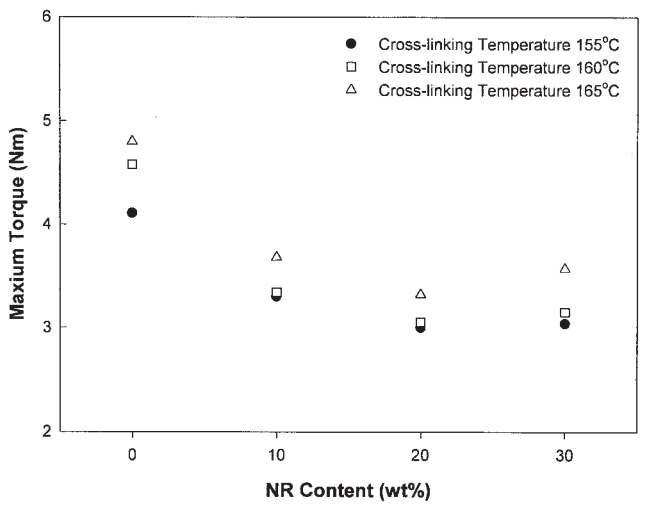

Figure 4 The variation of maximum torque of EVA/NR blend with blend composition at various temperatures.

TABLE II
Gel Content of EVA/NR Blend Foamed at 165°C

Composition	Gel content (%)
EVA 100	72.59
EVA90/NR10	57.68
EVA80/NR20	54.62
EVA70/NR30	46.63

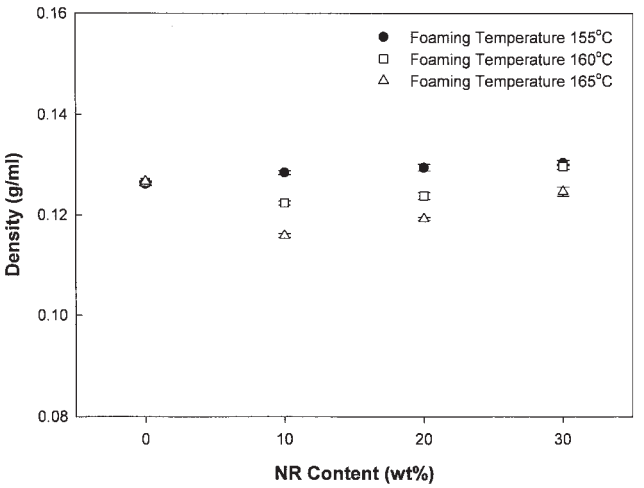

Figure 6 Change of density with EVA/NR foam composition at various temperatures.

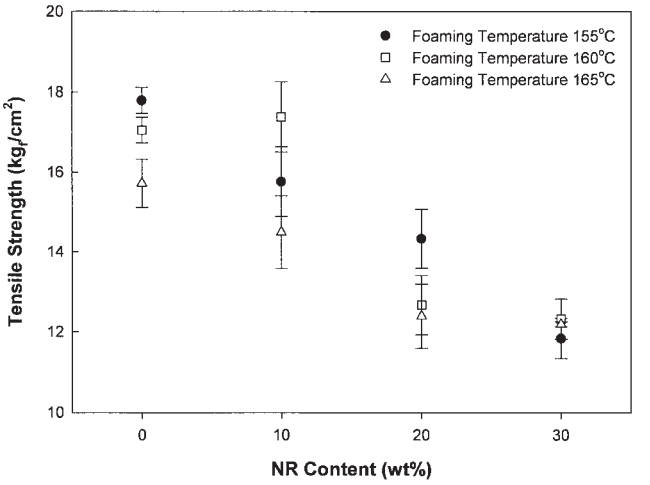

Figure 8 Tensile strength variation of foam with foam composition and foaming temperatures.

of NR in the blend, and the decrease of crosslinking temperature, decreases the crosslink density, and 165°C is the optimal temperature for crosslinking of EVA/NR blends.

The gel content (%) of EVA/NR blends foamed at 165°C was determined according to ASTM D2765–68 (method A). The crosslinked parts of EVA/NR foams, extracted using boiling xylene for 12 h, were dried and then weighed to estimate gel content. The gel content was found to decrease with increasing NR content (Table II).

According to the study conducted in our lab, the lowest foam density for EVA was achieved at 155°C. However, the lowest foam density for EVA/NR blends was not achieved at 155°C. The density of the EVA/NR blend foamed at 155°C is higher than the density of the EVA foamed at 155°C (Fig. 6). But the density of the EVA/NR blend foamed at 160°C and 165°C is lower than the density of the EVA foam. From this result, it is obvious that the control of crosslinking is very important for foaming. 165°C is the optimal temperature for a crosslinking of EVA/NR blends. As a result, the lowest foam density for EVA/NR blends is achieved at 165°C. An optimally crosslinked polymer expands, without rupture, to the greatest volume possible. Foaming at higher temperature, for example, 170°C was attempted; however, the result did not show any further reduction in foam density. Considering only the density, 90/10 (EVA/NR) and 165°C are the optimal composition and foaming temperature, respectively.

With increasing NR composition, rebound resilience of the EVA/NR foam increases (Fig. 7). Only with 10% addition of NR is the increase of rebound resilience

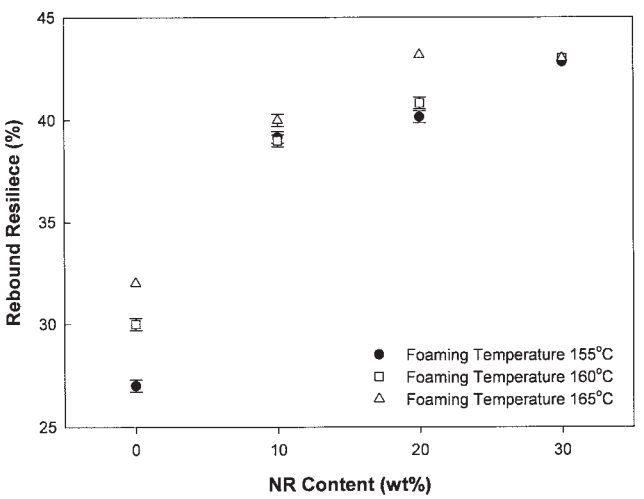

Figure 7 The change of rebound resilience with composition of EVA/NR foam and temperatures.

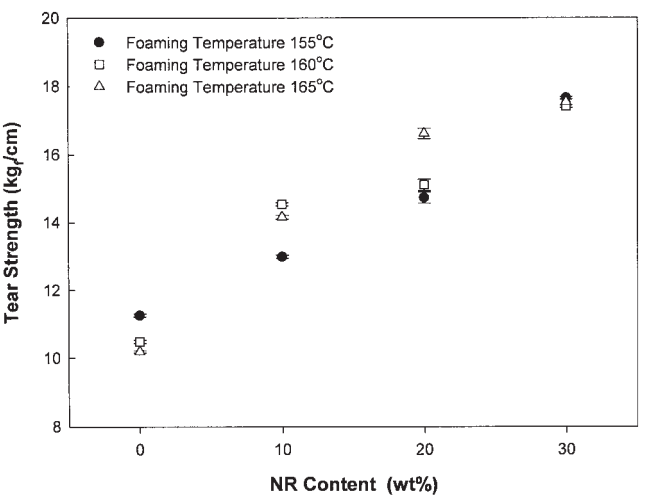

Figure 9 Variation of tear strength of EVA/NR foam with composition at various temperatures.

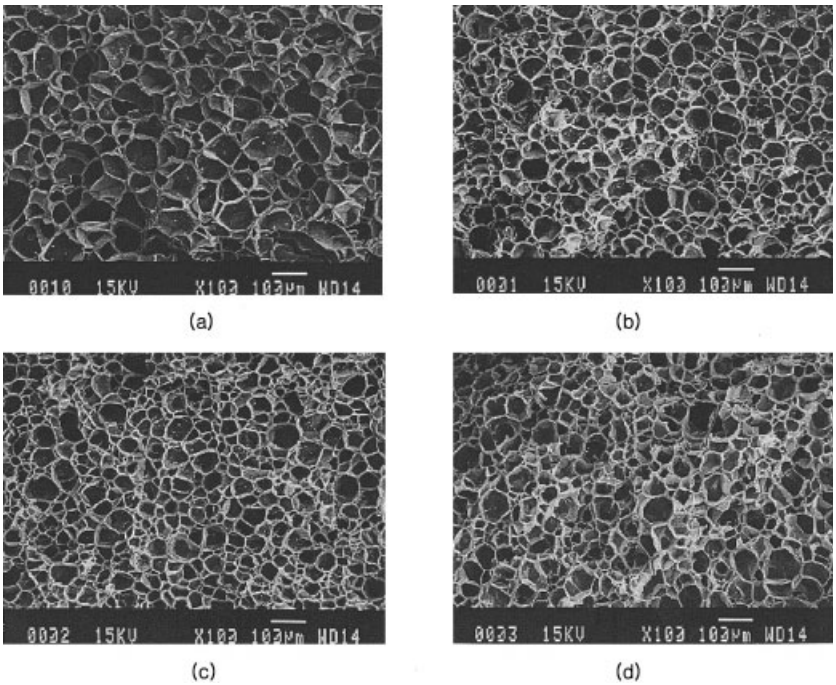

Figure 10 SEM photographs of EVA/NR foams with various composition: (a) EVA100, (b) EVA90/NR10, (c) EVA80/NR20, and (d) EVA70/NR 30.

significant. At the same blend ratio, rebound resilience at 165°C is slightly higher than at 155°C and 160°C.

Figure 8 shows the tensile strength of EVA/NR foams. With increasing NR composition, tensile strength decreases linearly. However, tear strengths of EVA/NR foams increase with increasing NR composition (Fig. 9). From this study, it was observed that EVA/NR (90/10) blend, foamed at 165°C, displayed lower density, better rebound resilience, and greater tear strength than EVA foam. A typical image of the cellular structure of the EVA/NR blend foamed at 155°C is shown in Figure 10. The EVA/NR foams have a closed-cell structure.

CONCLUSION

The density of EVA/NR foam foamed at 155°C is higher than the density of EVA foam foamed at 155°C, but the densities of EVA/NR foamed at 160°C and 165°C are lower than the density of the EVA foam. From this result, it is obvious that the control of crosslinking is very important for foaming. The optimal temperature for crosslinking of EVA/NR foam is found to be 165°C and, as a result, the density of EVA/NR foamed at 165°C is the lowest. It was also observed that EVA/NR (90/10) foamed at 165°C showed lower density, improved rebound resilience, and greater tear strength compared with EVA foam.

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