Swelling Behavior of NR/EPDM Rubber Blends Under Compression Strain

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ABSTRACT: The effect of different blend compositions of natural rubber (NR)/ethylene–propylene diene monomer rubber (EPDM) on the swelling behavior of that blend in motor oil under compression strain was investigated. The compression recovery of all blend ratios investigated had positive values at low applied compression values (3%). However, at high compression values (18 and 35%), the compression recovery had negative values. The lower weight uptake of motor oil was shown by the EPDM vulcanizate, whereas the 25/75 NR/EPDM blend showed the highest compression recovery. © 2001 John Wiley & Sons, Inc. J Appl Polym Sci 82: 3052–3057, 2001

Key words: rubber; blends; swelling; compression; elastomers

INTRODUCTION

The chemical or physical blending of two or more polymers is the simplest mean to obtain a variety of physical and chemical properties from the constituent polymers. There is no doubt that the main reason for blending is economy.[1] However, the gain in new properties depends on the degree of compatibility of a polymer at a certain molecular level. The compatibility of polymer blends has been reported, both experimentally and theoretically, including methods of determining the degree of compatibility.[2]

The swelling behavior of rubber blend vulcanizates is a diffusion process. As the diffusion process proceeds, the dimension of the rubber component increases until the concentration of the liquid is uniform throughout the component and an equilibrium state is achieved.[3] The amount of a given solvent that will diffuse into rubber until it reaches equilibrium swelling depends on the degree of compatibility of two polymers, the percentage of applied compression, and other factors that may be out beyond the scope of this study. Flory and Rehnner[4] showed that the volume uptake of a liquid by a rubber vulcanizate was altered by the application of stress. However, Trealor[5] gave a theory for the effect of applied stress on the swelling of a polymer and related the changes in the volume uptake, compression stress, and recovered length to the amount of applied compression, network breakdown, and the network of the rubber blend vulcanizate. Lawandy et al.[6–8] investigated the swelling behavior of polychloroprene rubber vulcanizate under compression strain. Other authors have studied the mechanical properties and morphology of polymer blends.[9,10] However, limited studies have been reported about the swelling of rubber blends in motor oil under different compressions.[11,12] The objective of this study was to investigate the swelling behavior of natural rubber (NR)/ethylene–propylene diene monomer rubber (EPDM) blends under compression strain. Another aim was to supply the oil-seal engineering manufacturer with information that may help in the se-

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lection of the proper blend ratio for an oil seal that can achieve better compression recovery when the stress applied on the seal is released.

EXPERIMENTAL

Materials

SMR-10 NR was produced in Malaysia. Vistalon 6505 EPDM was a product of Esso Chimie. Esso (London, UK) extra-multi-grade motor oil (20W-50AP/SF/CC) was a product of Exxon (Houston, TX, USA). Maleic anhydride (Merck, Munich, Germany) was used as a modifier for EPDM in the presence of a benzoyl peroxide initiator (Merck).

Techniques

Maleated EPDM was used as a compatibilizer for NR and EPDM. It was prepared by the addition of maleic anhydride (2 g) and benzoyl peroxide (0.15 g) to EPDM (100 g) on an open laboratory two-roll mill at 80°C. Different blend ratios of NR/EPDM were prepared. The ratios were 100/0, 75/25, 50/50, 25/75, and 0/100. The blend mixes were cured in a hydraulic press at the same temperature and time estimated with a Monsanto R-100 oscillating disc rheometer (Akron, OH) according to ASTM Standard D 2084-95. The base recipe of the NR/EPDM blends contained, in parts per hundred parts of rubber (phr), maleated EPDM (10 phr), zinc oxide (5 phr), stearic acid (2 phr), HAF-N330 (high-abrasion furnace black; 40 phr), processing oil (5 phr), sulfur (2.5 phr), and *N*-cyclohexyl 2-benzothiazole sulfenamide (1 phr).

Swelling and Compression Recovery Measurements

To study the effect of compression stress on the equilibrium swelling and compression recovery, we molded cylindrical samples 13 mm in diameter and 5 mm high. The samples were vulcanized under the same conditions of pressure, temperature, and time, as estimated with a Monsanto rheometer. We used a compression set clamp device to accommodate the five mixes at the same time. The clamp consisted of five circular, highly polished steel plates between which the test pieces were compressed. The plates were held together by a bolt. We placed spacers in the form of rings around the axial bolt and between the plates to limit the degree of compression of the test pieces. The spacers used were of different thicknesses. The procedure used in these measurements can be summarized as follows:

1. The test pieces were marked and placed symmetrically between the plates of the compression device after their weight was accurately measured in air. The exact thicknesses of the samples were measured with a rubber thickness gauge; the thicknesses were measured to the nearest 0.01 mm.
2. The bolt was tightened uniformly until the plates were in contact with the spacers, and then the compression set device was immersed completely in motor oil. The oil container was then transferred to an air-circulating oven fixed at 100°C.
3. After each time interval, the clamp device was parted, and the excess oil on the surface of the test pieces was removed by plotting with filter paper; then, the test pieces were allowed to recover for 20 min.
4. Their thickness and weight were measured accurately, and the samples were reimmersed in oil. This procedure was repeated until the samples reached equilibrium swelling.

The difference between the thickness after recovery (h_r) and the original thickness (h_o) of the test piece is expressed as a percentage of the initially applied compression, where h_s is the spacer thickness. This can be related as

$$\%\text{ compression recovery} = [(h_r - h_o)/(h_o - h_s)] \times 100$$

The weight uptake is given by the following equation, where W_s and W_o are the weights of a specimen after and before swelling in motor oil, respectively:

$$\text{The weight uptake }(\%) = [(W_s - W_o)/W_o] \times 100$$

RESULTS AND DISCUSSION

To investigate the diffusion behavior of motor oil through NR/EPDM rubber blend vulcanizates under compression strain, we subjected vulcanizates with different blend ratios, as indicated, to swelling in motor oil under different compression strains (3, 18, and 35%) at 100°C. The weight uptake percentage was determined and plotted against the square root of the swelling exposure time (minutes) in motor oil (Figs. 1–3). The weight uptake increased as the exposure time

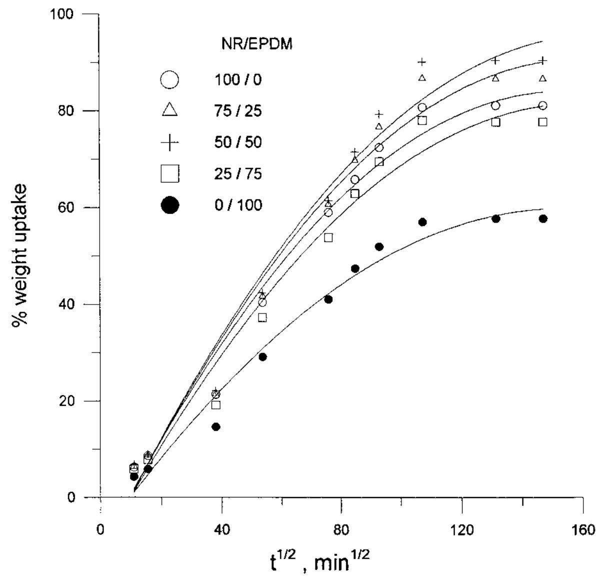

Figure 1 Weight uptake (%) of motor oil for NR/EPDM vulcanizates versus the exposure time ($t^{1/2}$) at 100°C and 3% compression.

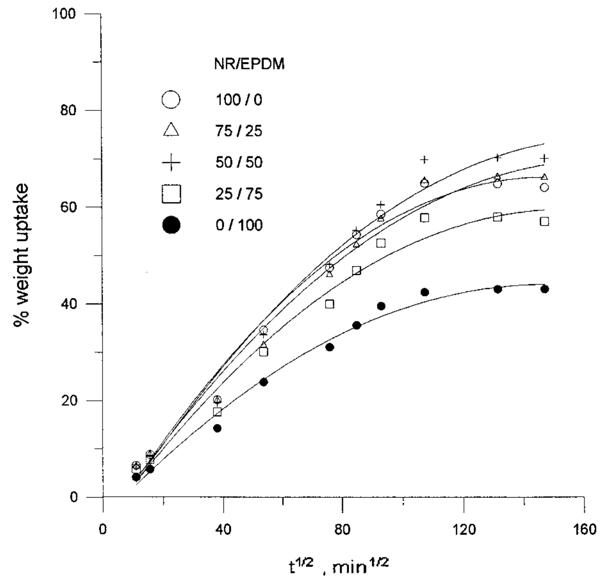

Figure 3 Weight uptake (%) of motor oil for NR/EPDM vulcanizates versus the exposure time ($t^{1/2}$) at 100°C and 35% compression.

increased, and equilibrium swelling was achieved after 192 h of immersion in oil. The time for equilibrium swelling was the same for all compression percentages applied. The swelling commenced with the absorption of liquid in the surface layer of the rubber sample to a certain concentration. Then, the swelling proceeded with the depth of the swollen layers increasing until the sample reached equilibrium swelling. Also, the time taken to reach equilibrium swelling was independent of the blend ratios investigated. The lowest weight uptake was recorded with a mix containing EPDM polymer only (0/100), whereas the 25/75 NR/EPDM blend ratio recorded a lower weight uptake value. Generally, Figures 1–3 show that the curves follow the same pattern. It is also noted that the curve levels depended on the degree of compression applied.

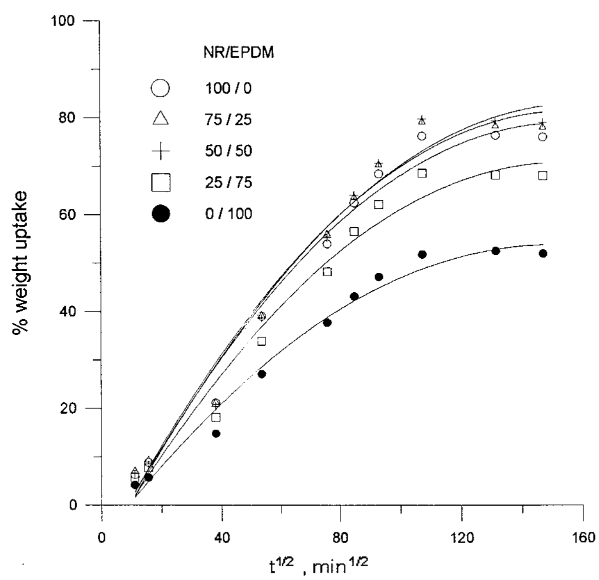

Figure 2 Weight uptake (%) of motor oil for NR/EPDM vulcanizates versus the exposure time ($t^{1/2}$) at 100°C and 18% compression.

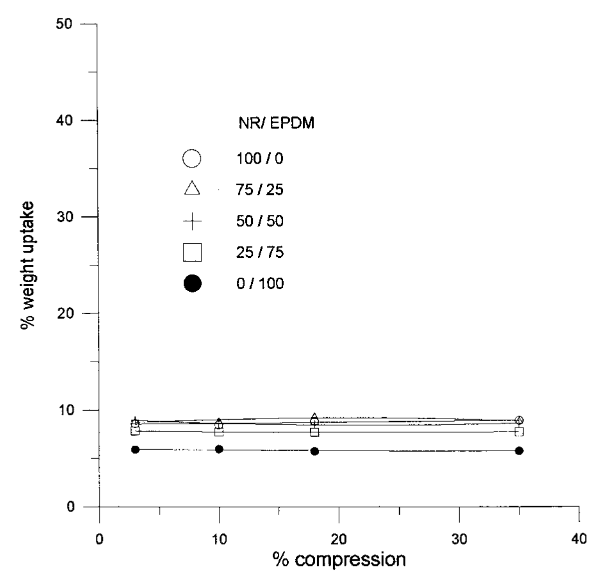

Figure 4 Weight uptake (%) of motor oil for NR/EPDM vulcanizates versus compression (%) for 4 h at 100°C.

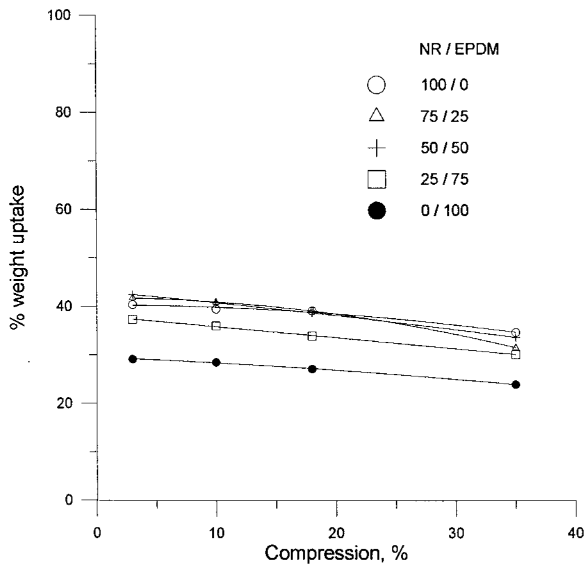

Figure 5 Weight uptake (%) of motor oil for NR/EPDM vulcanizates versus compression (%) for 48 h at 100°C.

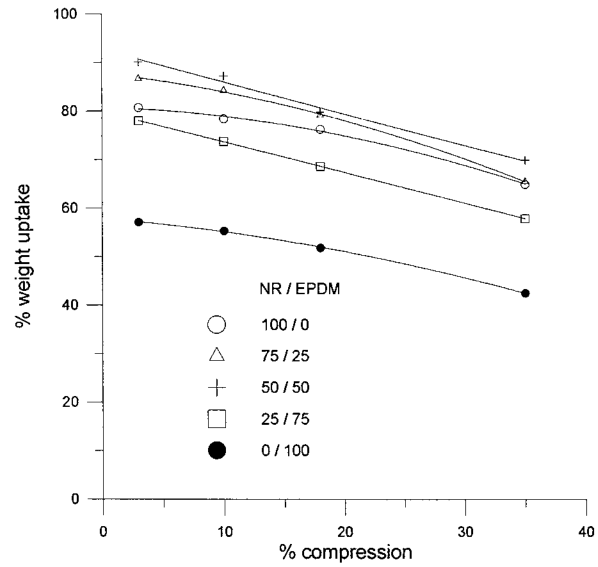

Figure 7 Weight uptake (%) of motor oil for NR/EPDM vulcanizates versus compression (%) for 192 h at 100°C.

To study the effect of compression on the swelling behavior of NR/EPDM blend vulcanizates, at different exposure times in motor oil, we estimated the weight uptake and plotted it against the compression percentage for different periods (4, 48, 120, and 192 h). This is shown in Figures 4–7. It is clear that, for a short exposure time of 4 h (Fig. 4), no significant change in the weight uptake was recorded, and the weight uptake was independent of the compression. However, at the relatively longer exposure time of 48 h (Fig. 5), the weight uptake began to be affected slightly with the compression, whereas at the long exposure times of 120 and 192 h (Figs. 6 and 7), the decrease in the weight uptake with the increase in compression, was much more pronounced.

The compression recovery of the swelled rubber vulcanizates was determined and plotted versus

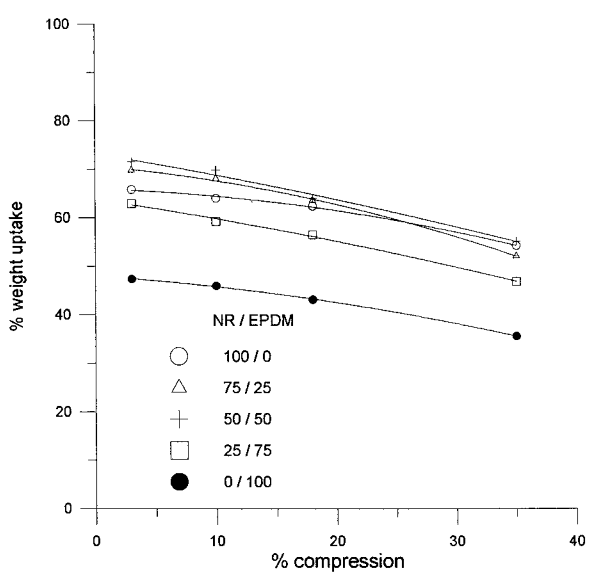

Figure 6 Weight uptake (%) of motor oil for NR/EPDM vulcanizates versus compression (%) for 120 h at 100°C.

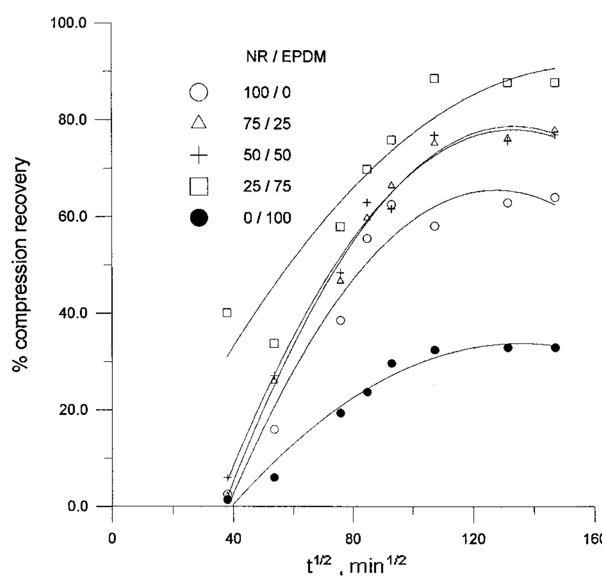

Figure 8 Compression recovery (%) of NR/EPDM vulcanizates versus the exposure time ($t^{1/2}$) in motor oil at 100°C and 3% compression.

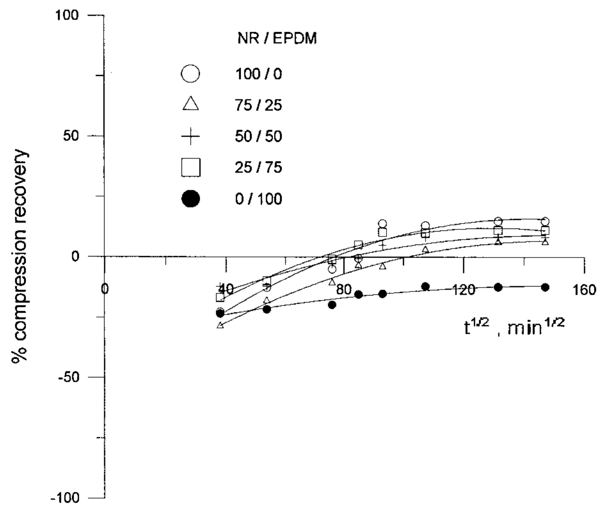

Figure 9 Compression recovery (%) of NR/EPDM vulcanizates versus the exposure time ($t^{1/2}$) in motor oil at 100°C and 10% compression.

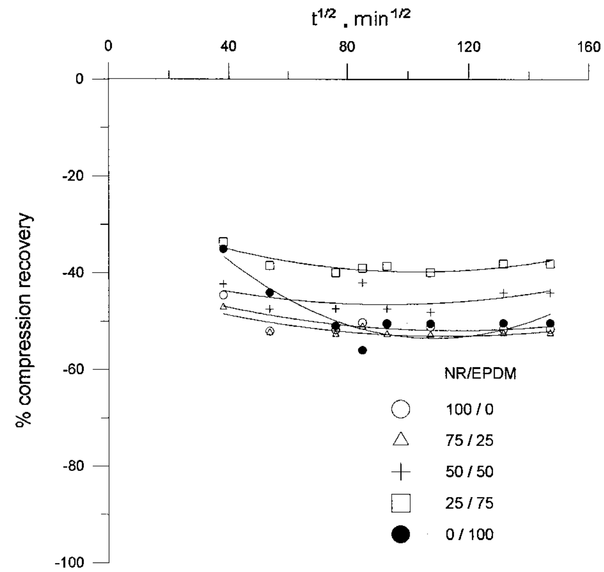

Figure 11 Compression recovery (%) of NR/EPDM vulcanizates versus the exposure time ($t^{1/2}$) in motor oil at 100°C and 35% compression

the square root of the swelling exposure time. This is in Figures 8–11. Each figure represents the recovery at a certain percentage of compression and contains five curves describing the behavior of rubber blends at different exposure times. At low compression (3%), the recovery was positive (Fig. 8). This recovery was a function of the swelling, which varied with the blend ratio. The lowest recovery values were recorded with the EPDM (0/100) vulcanizate. However, the 25/75 NR/EPDM blend ratio showed the highest recovery values. The recovery increased extensively with the swelling exposure time and reached a steady state after 192 h. At higher compression (10%), the recovery curves showed negative and positive values, depending on the exposure time (Fig. 9). At a low exposure time, the recovery percentage was negative where the rubber vulcanizates were considered to be subjected to the compression force only. However, as the swelling proceeded, the recovery was a function of two forces, the swelling pressure and the com-

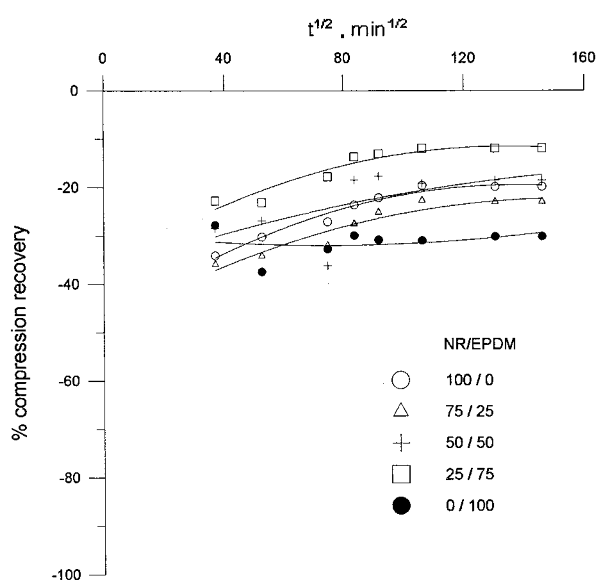

Figure 10 Compression recovery (%) of NR/EPDM vulcanizates versus the exposure time ($t^{1/2}$) in motor oil at 100°C and 18% compression.

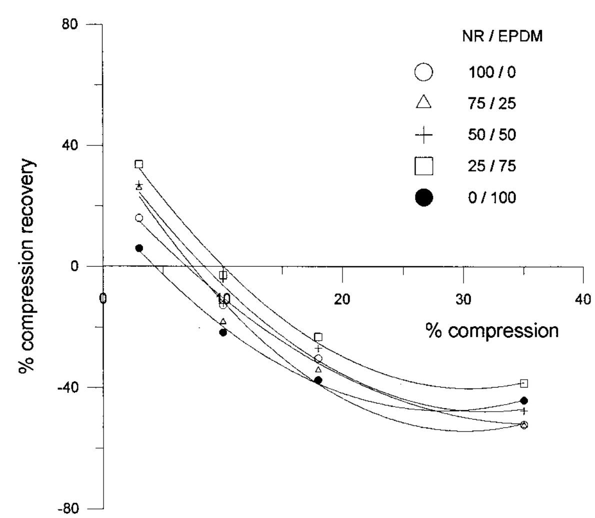

Figure 12 Compression recovery (%) of NR/EPDM vulcanizates with different blend ratios versus compression (%) in motor oil for 48 h at 100°C.

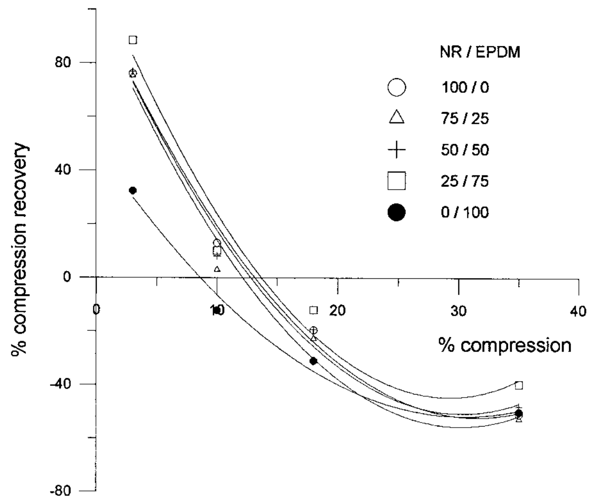

Figure 13 Compression recovery (%) of NR/EPDM vulcanizates with different blend ratios versus compression (%) in motor oil for 192 h at 100°C.

pression force; as a result, the recovery showed positive values at longer exposure times. At higher compressions (18 and 35%), the recovery curves showed only negative values, as shown in Figures 10 and 11. These negative values were also the result of two forces, the compression force and the swelling force in the opposite direction. The recovery at 18% compression (Fig. 10) increased slightly with increasing exposure time. However, at 35% compression (Fig. 11), the recovery remained unchanged after 96 h of exposure time for all the NR/EPDM vulcanizates under investigation. The 25/75 NR/EPDM blend ratio showed the highest compression recovery at all compressions applied.

The compression recovery percentage was plotted against the applied compression percentage (Figs. 12 and 13) at 48 and 192 h of exposure time. The recovery decreased with increasing applied compression. That behavior was the same regardless of the length of the exposure time. The 25/75 NR/EPDM ratio showed the highest degree of elastic recovery.

CONCLUSIONS

1. The time taken to reach equilibrium swelling, in motor oil, of the NR/EPDM vulcanizate was independent of the blend ratios investigated. The lowest weight uptake was recorded with the individual EPDM vulcanizate (0/100). However, the 25/75 NR/EPDM blend showed the lowest weight uptake among the other blend ratios, including the NR vulcanizate.
2. At a short exposure time in motor oil, the weight uptake was independent of the applied compression, whereas at longer exposure times, a decrease in the weight uptake with the applied compression was much more pronounced.
3. At low compression (3%), the compression recovery percentage for all blend ratios was positive. At high compression, the recovery had negative values. The highest recovery value was recorded for the 25/75 NR/EPDM blend. However, the lowest recovery was observed with the EPDM vulcanizate (0/100).
4. The compression recovery decreased with increasing applied compression for all exposure periods.
5. The 25/75 NR/EPDM blend was advantageous because of its high degree of elastic recovery and its lower weight uptake of motor oil.

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