Development of an enabling EPDM polymer to improve performance and processability of EPDM compounds

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Royalene EPDM, a terpolymer of ethylene, propylene and a non-conjugated diene, is one of the most versatile specialty elastomers in commercial use today. The main attributes of EPDM are its outstanding resistance to oxidation, ozone and the effects of weathering. It is also highly extendable, allowing high levels of fillers and plasticizers to be added, while still maintaining good physical properties. Its low specific gravity, combined with its high extendability, allow for economical functional parts to be produced. Royalene EPDM, the "crackless rubber," has found wide use in applications that take advantage of its excellent ozone crack resistance and heat aging characteristics, as well as its low temperature flexibility, chemical resistance and electrical properties.

The global EPDM market size is currently estimated to be about 1,450 kilotons, and is expected to be about 1,600 kilotons by 2023. Asia Pacific is the largest regional market, with over 40% of the global demand, followed by the Americas and Europe. Key application areas include automotive, single ply roof membranes, thermoplastic elastomers, wire and cable insulation and oil additives. Automotive related applications such as weatherseals, hoses and tire sidewalls represent the largest application area in major regional markets (ref. 1).

Lion Elastomers' EPDM products are manufactured with a Ziegler-Natta catalyst in a solution process which enables the manufacturing of polymers with different molecular weights, all the way from ultra high molecular weight with oil extension to very low molecular weight, supplied in liquid form and marketed as Trilene liquid EPDM (refs. 2 and 3). The major applications for Trilene liquid EPDM are adhesives, non-extractable plasticizers and polymeric process aids. Lion has pioneered the use of EPDM in new applications leading to the introduction of Trilene liquid EPDM polymers for the coatings industry (ref. 4). Trilene 65, a low molecular weight EPDM terpolymer, provides coatings formulators with strong adhesion to a variety of substrates, excellent durability under the harshest environmental conditions, and superior impact resistance and flexibility at low temperatures. Trilene 65 can be formulated into high solids, VOC compliant coatings that provide the performance of traditional EPDM materials. In the last couple years, Lion Elastomers developed an aqueous dispersion of Trilene 65 called Trilene 65D. The aqueous dispersion of Trilene 65D allows manufacturers of coatings to formulate waterborne coatings to minimize the VOCs in their products (refs. 5-7).

Due to concerns over global warming caused by man-made activities such as CO_2 emissions from internal combustion vehicles (ICV), the regulation of fuel efficiency (CO_2 emissions) of automobiles is expected to be strengthened by emerging countries in ways similar to those driven by the industrialized countries of the EU and U.S. When fuel efficiency (CO_2 emissions) regulation is strengthened to about 60 to 70 g/km, each global automotive manufacturing company will be required to have electric vehicles (EVs) and plug-in hybrid electric vehicles (PHEVs). Major global automakers have ambitious plans to introduce EVs and PHEVs to the marketplace by 2025 (ref. 8). Such a change will lead to the possibility that existing parts makers may lose their businesses entirely (e.g., engine components, drive units, transmissions and control devices). The industry structure will also change from vertical integration to horizontal specialization (ref. 9). Electrification is shaking up the automotive parts supply chain, forcing rubber product companies to add new capabilities to their established areas of expertise in materials science and technology (ref. 10). The NVH (noise, vibration and harshness) issues of automobiles are shifting from the vibrations caused by internal combustion engines to the vibrations and noise caused by motors and gearboxes (ref. 11). With the noise produced by traditional combustion engines either intermittent or eliminated entirely, noise from the vehicle transmission and driveline is an area of greater interest as a larger proportion of the overall vehicle noise. There is no need for high heat resistant EPDM polymers in under-the-hood applications such as engine mounts, coolant hoses, power transmission belts, muffler hangers, etc., in the EVs. On the other hand, opportunities exist for high Mooney EPDM polymers in automotive weather seals, as there is a need for better vehicle sealing systems to isolate the noises (road, wind, drive chain, etc.) from outside the passenger compartment while the vehicles are running at high speed. There is also a need to reduce the weight of the automotive parts, including the weather seals, while making them better for noise isolation, which would require ultra high MW EPDM polymers that can deliver higher performance without losing processability.

In recent years, Lion Elastomers has developed a series of Royalene EPDM grades by employing a novel, newly developed catalyst package, resulting in a unique molecular architecture which has the characteristics of both long chain branching (LCB) and bi-modal molecular weight distribution (MWD) based on the high temperature size exclusion chromatograph (SEC) analyses (ref. 12). An ultra premium, high performance sponge and dense extrusion grade Royalene 636 was one of these new EPDM grades, which was later replaced with a non-oil extended version, Royalene 515 (ref. 13). These Royalene grades were specifically designed for production of best-in-class, ultra premium, high performance sponge and dense extrusions. The attributes of Royalene 515 include: very high diene content for ultra fast cures and outstanding compression set resistance; high molecular weight for superior physical properties and compound cost reduction; high viscosity at low shear rates for excellent melt strength and shape retention; low

Table 1 - characteristics of ultra high molecular weight amorphous EPDM polymer Royalene 5169

ML 1+8 at 150°C, MU	120
E/P weight ratio	62/38
Ethylene, weight %	58.6
ENB, weight %	5.5
Oil content, phr	19
MWD	Medium (unique)
Physical form	Friable bale

viscosity at high shear rates for superior processing at high extrusion speeds; Class A surface quality on both sponge and dense extrusions; and they are offered as easy to mix friable bales. With the same process and catalyst technology, a new high performance EPDM polymer with improved load bearing dynamic properties, Royalene 5158, was developed for automotive anti-vibration applications that require high tensile strength, low compression set, low damping and a low dynamic-to-static spring ratio (ref. 14).

In 2018, Lion Elastomers introduced the fourth EPDM product with the same polymer attributes, an ultra high molecular weight amorphous EPDM polymer: Royalene 5169 (ref. 15). The purpose of developing this polymer was to create an ultra high molecular weight, amorphous EPDM polymer that meets customers' cost competitive and processing needs, such as highly extended formulations with good processability, low temperature compression set properties and availability in a friable and easy to mix bale form. The Royalene 5169 polymer has been successfully formulated as a sole EPDM polymer or as an enabling additive for other EPDM polymers in compounds for several intended applications such as highly filled extrusion profiles, coolant hose, mechanical goods (low durometer compounds), wire and cable, TPV and anti-vibration products by customers worldwide since its introduction in the EPDM market. The characteristics of the Royalene 5169 EPDM polymer are shown in table 1.

Comparison of friability of Royalene 5169 with semi-crystalline Royalene EPDM polymers

Friability is a desirable property of baled rubber products for mixing of rubber compounds, as it allows one pass mixing of compounds in internal mixers so that compound manufacturers can increase productivity in mixing operations and realize higher profitability with lower compound cost. Friability can be defined as "the ability of baled rubber to break down under shear to form particles substantially similar in size and distribution to that of the original particulate rubber" (ref. 16). Baled rubber with good friability will break down quickly into small particle sizes in the initial stage of mixing in the internal mixer after bales have been loaded into the mixer to allow quick incorporation of organic/inorganic fillers and plasticizers, and will reduce the BIT (black incorporation time), which is the critical step in the rubber compound mixing cycle. Lion Elastomers has developed a proprietary "friability" test in which a shear force is applied to baled rubber samples at a constant rate for a fixed period of time. The test procedure was designed to duplicate the initial stage of baled rubber in an internal mixer after it has been loaded into the mixer. After the test is completed, the rubber samples are collected and screened using screens of different mesh sizes. This test was conducted on the new amorphous Royalene EPDM grades 5169 and 515, and compared with the well known friable semi-crystalline Royalene EPDM grades 510, 539 and 556, which have been used by rubber compound manufacturers in the one-pass mixing process successfully for years. The test results from these Royalene products are shown in figure 1, compared with a densified rubber bale. The test results clearly show that both the high Mooney amorphous Royalene 5169 and 515 EPDM grades can be broken down into small particle sizes like the lower Mooney semi-crystalline EPDM grades under the same shear conditions. The densified non-friable bale had about 30% by weight particles with sizes greater than 12.7 mm (0.5 inch), while all the other Royalene EPDM bales were friable, as they all had less than 10% by weight particles with sizes greater than 12.7 mm. In fact, the weighted average particle size of both Royalene 5169 and 515 was about 3 mm, while that of Royalene 510, 539 and 556 was 4 mm, and that of the densified non-friable bale was 8 mm. This friability test result was also validated in Lion's own laboratory mix-

Figure 1 - comparison of particle size distribution of Royalene 5169, 515 and semi-crystalline Royalene EPDM polymers after the friability test

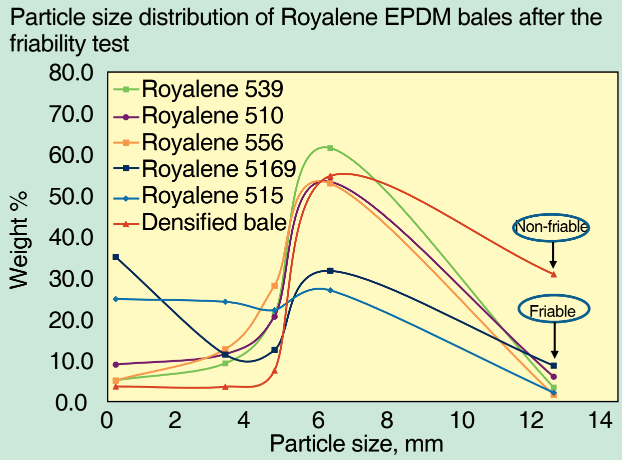


Table 2 - characteristics of comparative EPDM polymers

EPDM polymer	*Mooney viscosity, ML1+4 at 125°C*	*E/P ratio*	*Ethylene, wt.%*	*ENB, wt.%*	*Oil extension, phr*	*MWD*
Royalene 563	75	60/40	57.3	4.5	0	Medium
Royalene 515	115	62/38	56.1	9.5	0	Medium (unique)
Polymer A	122	56/44	52.4	6.5	0	Very broad (bimodal)
Polymer B	127	62/38	58.6	5.4	15	Broad (CLCB)

Table 3 - formulations of low, medium and high filler loading compounds with sulfur cure

Formulation (phr)	*Low*	*Medium*	*High*
Polymer	100 + X	100 + X	100 + X
N-650 black	145	-	-
N-550 black	-	180	205
Calcium carbonate	-	55	150
Paraffinic oil	100 - X	135 - X	150 - X
Zinc oxide	5	5	5
Stearic acid	1	1	2
Sulfur	0.1	0.1	0.1
Paraffin wax	-	-	2
Masterbatch total	*351.1*	*476.1*	*614.1*
MBT	0.5	-	-
MBTS	0.5	3	3
ZDBC	1	2.5	2.5
TMTD	0.5	0.8	0.8
DPTH	0.5	0.8	0.8
Sulfur	0.9	0.4	0.4
Total	*355*	*483.6*	*621.6*

X: Oil content in the EPDM product

ing and extrusion evaluations, and in customers' commercial scale mixing and extrusion operations.

Evaluation of Royalene 5169 in application formulations

Royalene 5169 EPDM polymer was evaluated in several application formulations in Lion's applications laboratory versus high Mooney amorphous Royalene EPDM grades 563 and 515, and a couple of comparative EPDM grades from other EPDM manufacturers. The properties of these EPDM polymers are shown in table 2, while the dynamic mechanical properties of these polymers as tested on a rubber process analyzer (RPA 2000, Alpha Technologies) are shown in figures 2 and 3. The graphs of complex viscosity and tangent delta versus frequency showed that Royalene 563 had a more linear structure and narrower MWD (molecular weight distribution) than the other EPDM polymers, while polymers A and B showed the characteristics of either bimodal MWD or CLCB (controlled long chain branching) structure (refs. 17 and 18). Royalene 5169 and 515 have characteristics of both bimodal MWD and CLCB structure.

Figure 2 - complex viscosity of Royalene 5169 and comparative EPDM polymers (rubber process analyzer, 150°C, 14% strain)

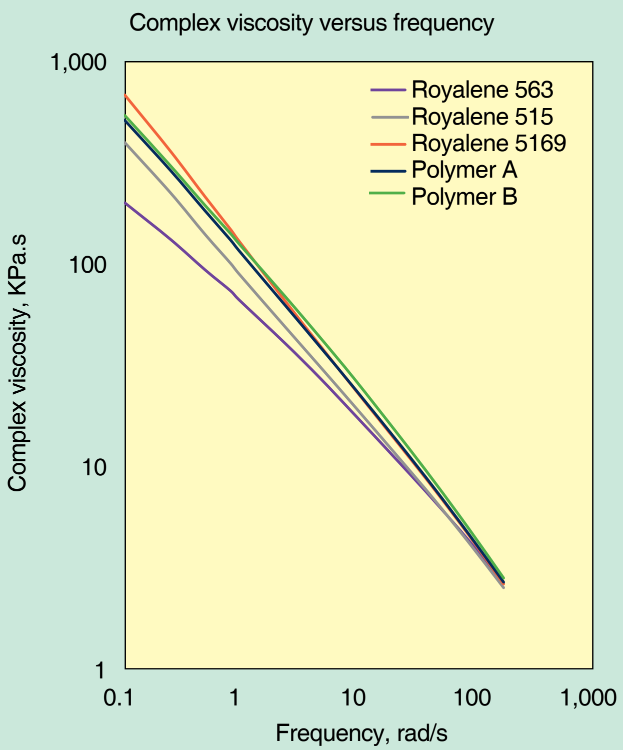
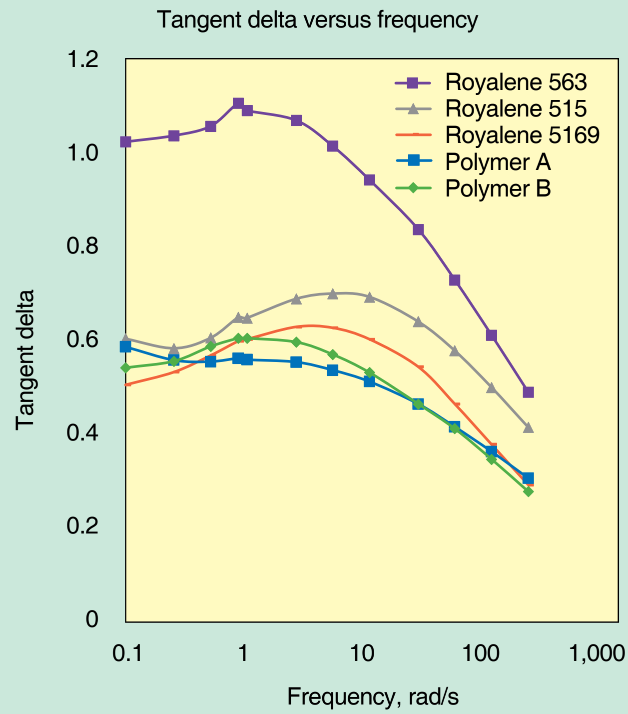


Figure 3 - tangent delta of Royalene 5169 and comparative EPDM polymers (rubber process analyzer, 150°C, 14% strain)



Royalene 5169 applications

Low/medium/high filler formulation (durometer 70 A, sulfur cure)

The competitive landscape continually pushes EPDM users to be more cost competitive using the same types of polymers for specific applications, and thus demands the same of their suppliers. The market demanded an amorphous EPDM product which can be highly filled while maintaining its processability and low temperature compression set properties. Table 3 shows the durometer 70 A compound formulations with different filler/oil loadings (low: 145/100, medium: 235/135 and high: 355/155) for comparison of Royalene 5169 with the high Mooney amorphous EPDM polymers shown in table 2. The rheological and cured physical properties of these compounds

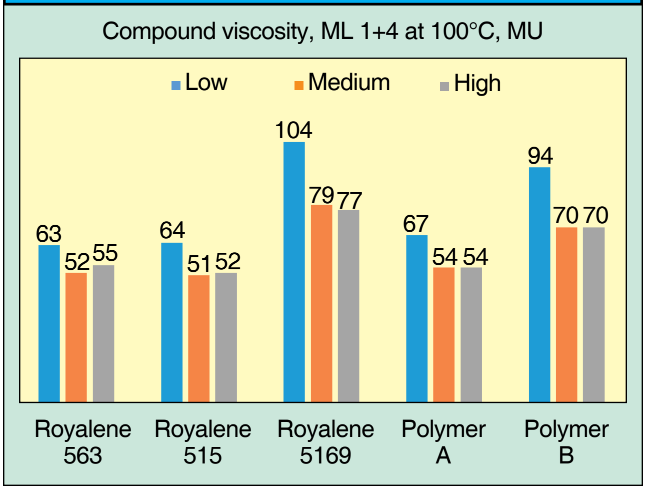


Figure 4 - compound viscosity of Royalene 5169 and comparative EPDM polymers in low, medium and high filler formulations

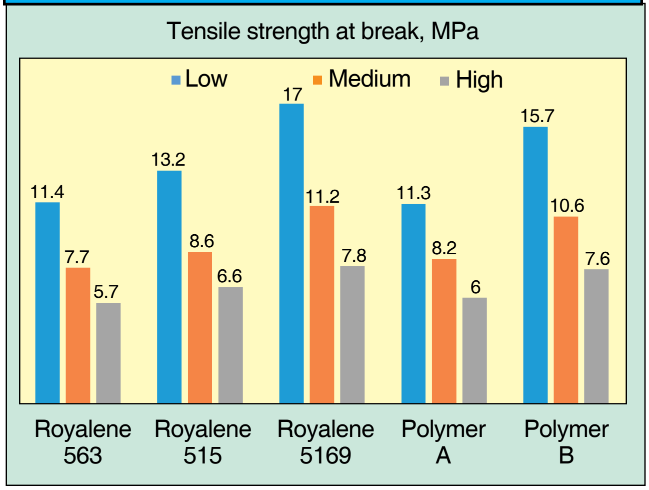


Figure 6 - tensile strength of Royalene 5169 and comparative EPDM polymers in low, medium and high filler formulations

are shown in figures 4-10. In figure 4, at the low filler/oil loading, the EPDM polymers with ultra high molecular weight, Royalene 5169 and polymer B, have very high compound viscosity at 104 MU and 94 MU, respectively, due to their high molecular weights, while the other three EPDM polymers with lower molecular weight have comparable compound Mooney at about 65 MU. At higher filler/oil loadings, the compound Mooney of both Royalene 5169 and polymer B dropped to the 70s, while the compound Mooney of the other three polymers dropped to the 50s, and there was almost no difference in the compound Mooney between the medium and high filler/oil loadings for all the polymers.

The crosslink density as measured by MH-ML in the cure rheometer test is shown in figure 5. In figure 5, at the low filler/oil loading, Royalene 515 has the highest crosslink density (26 dNm) because of its high ENB content. Royalene 5169, polymer A and polymer B have high crosslink density in the range of 20-22 dNm, while Royalene 563 has the lowest crosslink density because it has both low molecular weight and ENB content. At higher filler/oil loadings, the crosslink density of Royalene 515 is still the highest at 15-16 dNm, and Royalene 563 has the lowest crosslink density at about 10 dNm, while Royalene 5169, polymer A and polymer B have crosslink density at about 12-13 dNm; and there was almost no difference in the crosslink density between the medium and high filler/oil loadings for all the polymers.

In figure 6, at the low filler/oil loading, the Royalene 5169 and polymer B have the highest original tensile strength (17 MPa and 16 MPa, respectively), followed by Royalene 515 at about 13 MPa; while Royalene 563 and polymer A have lower tensile strength at about 11 MPa because they have lower molecular weight. At medium filler/oil loading, the tensile strength of both Royalene 5169 and polymer B stays above 10 MPa, while that of the other three polymers decreased to about 8 MPa. At the high filler/oil loading, the tensile strength of both Royalene 5169 and polymer B remains above 7 MPa, while that of the other three polymers decreased to about 6.5 MPa or lower.

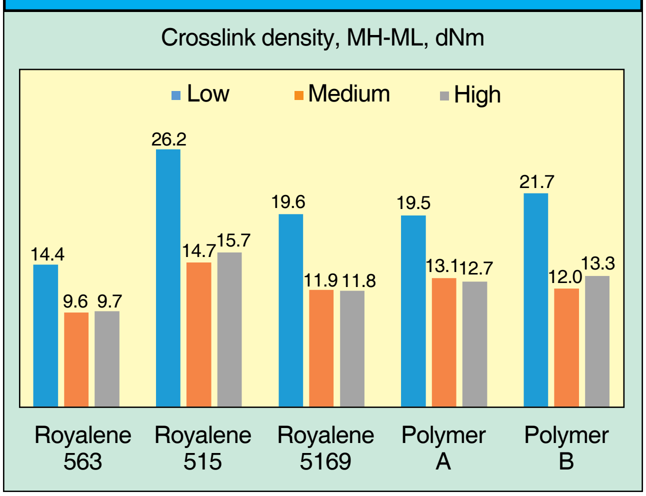


Figure 5 - crosslink density of Royalene 5169 and comparative EPDM polymers in low, medium and high filler formulations

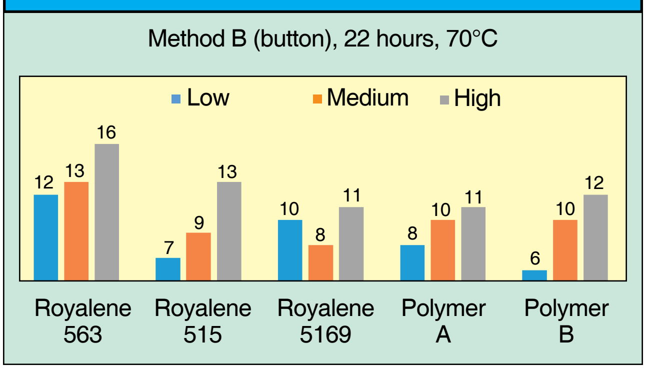


Figure 7 - compression set (22 hours at 70°C) of Royalene 5169 and comparative EPDM polymers in low, medium and high filler formulations

Figure 8 - compression set (70 hours at 125°C) of Royalene 5169 and comparative EPDM polymers in low, medium and high filler formulations

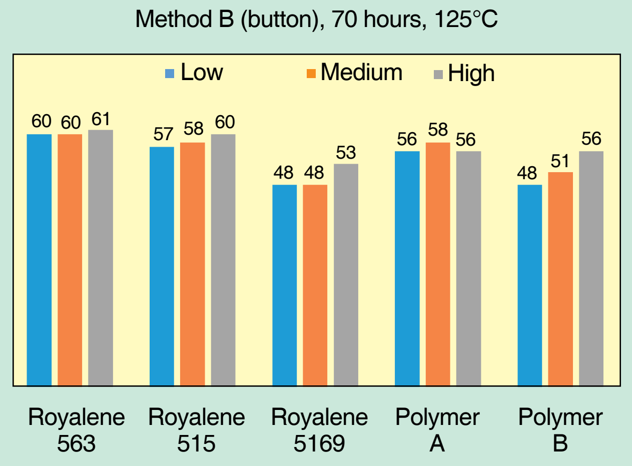
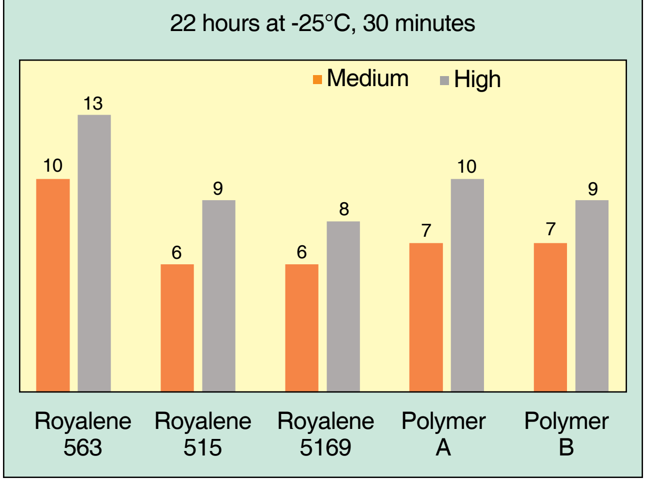
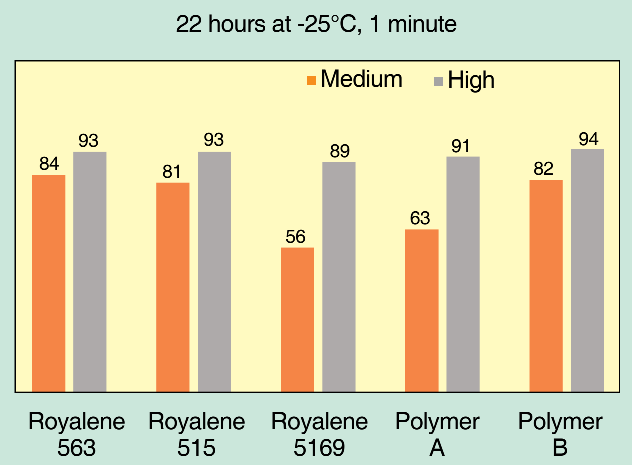


Figure 10 - compression set (22 hours at -25°C, 30 minute) of Royalene 5169 and comparative EPDM polymers in medium and high filler formulations



The compression set properties at elevated temperatures (22 hours at 70°C and 70 hours at 125°C) are shown in figures 7 and 8. The compression set for 22 hours at 70°C in figure 7 shows that all the EPDM polymers have comparable compression set properties in the range of 6% to 10% for both low and medium filler/oil loadings except Royalene 563, which has lower molecular weight than the other polymers. At the high filler/oil loading, the compression set is slightly higher in the range of 11% to 13%. However, the compression set for 70 hours at 125°C in figure 8 shows that Royalene 5169 and polymer B have the lowest compression set for both low and medium filler/oil loadings at 50% or less, while the other three polymers have compression set in the high 50s or low 60s as a result of their molecular weight. At high filler/oil loading, the compression set of Royalene 5169 increases slightly to the low 50s, while that of polymer B increases to the mid 50s.

Figure 9 - compression set (22 hours at -25°C, 1 minute) of Royalene 5169 and comparative EPDM polymers in medium and high filler formulations



The compression set at -25°C and one minute, shown in figure 9, shows that Royalene 5169 and polymer A with medium filler/oil loading have the lowest compression set at 56% and 63%, respectively, because they both have lower ethylene content than the other EPDM polymers; while the other three EPDM polymers have considerably higher compression set at about 80% or above. At high filler/oil loading, all the polymers have high compression set at -25°C and one minute at about 90%. When the compression set is measured after relaxing at room temperature for 30 minutes, as shown in figure 10, they all have comparable low temperature compression set properties in the range of 6% to 10% for both medium and high filler/oil loadings.

Rubber rich/low filler formulation (durometer 65 A, peroxide cure)

In many automotive under-the-hood applications, high heat aging resistance at 150°C or 160°C is required, such as for power transmission belts, coolant hoses, heat shield, etc. The peroxide cure systems are commonly used in these applications to meet these heat aging requirements. Table 4 depicts a durometer 65 A rubber-rich compound formulation with a peroxide cure system as an illustration of what impacts the differences in the polymer's molecular weight and ethylene content could have on the rheological and physical properties

Table 4 - rubber-rich compound formulation with peroxide cure

Formulation	*phr*
Polymer	100 + X
N-650 black	145
Paraffinic oil	100 - X
Zinc oxide	5
Zinc stearate	1
Antioxidant 405	1.5
Masterbatch total	*352.5*
TMPTMA coagent	2
DiCup 40KE peroxide	10
Total	*364.5*

X: Oil content in the EPDM product

Figure 11 - compound viscosity of Royalene 5169 and comparative EPDM polymers in low filler peroxide cure formulation

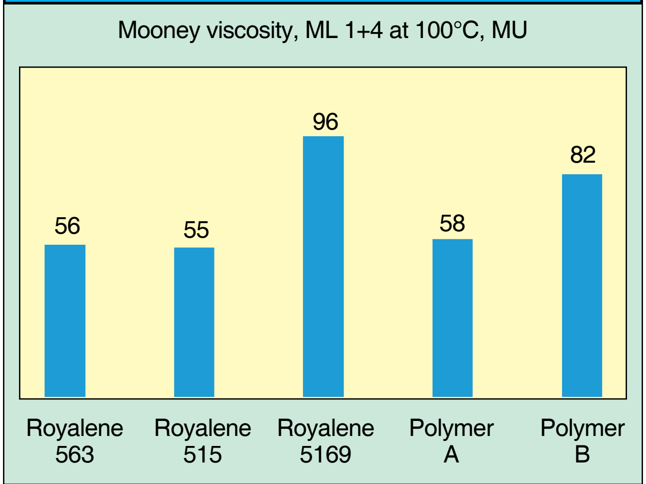
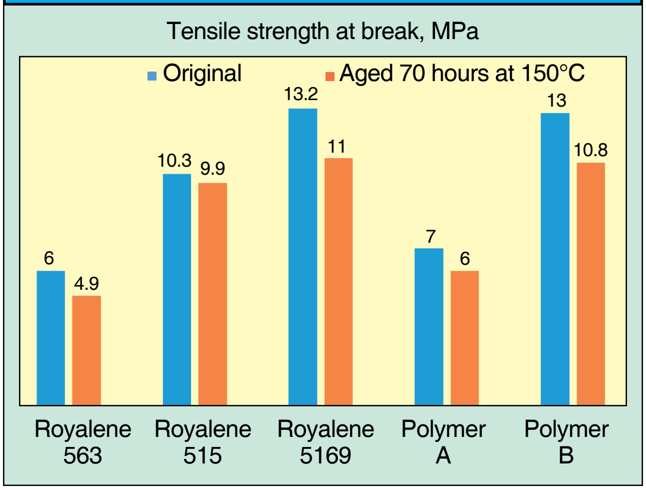


Figure 13 - original and heat aged tensile strength of Royalene 5169 and comparative EPDM polymers in low filler peroxide cure formulation



of the compounds. Figures 11-15 show the compound viscosity and the cured physical properties of these compounds. In figure 11, the EPDM polymers with ultra high molecular weight, Royalene 5169 and polymer B, have very high compound viscosity at 96 MU and 82 MU, respectively, due to their high molecular weights; while the other three EPDM polymers with lower molecular weight have comparable compound Mooney at about 55 MU. Figure 12 shows that the polymers with either high molecular weight (Royalene 5169 and polymer B) or high ENB content (Royalene 515) have high crosslink density (MH-ML), while the polymers with lower molecular weight (Royalene 563 and polymer A) have lower crosslink density. In figure 13, Royalene 5169 and polymer B have the highest original tensile strength (~13 MPa), followed by Royalene 515 at about 10 MPa; while Royalene 563 and polymer A have lower tensile strength because they have lower molecular weight. The heat aged (70 hours at 150°C) tensile strength of these polymers follows the same trend. The original and heat aged elongation at break are shown in figure 14, in which the Royalene 563 with the lowest crosslink density has the highest values of original and heat aged elongation at break, followed by polymer A; while the other three EPDM polymers with the highest crosslink density have about the same elongations at break. The compression set at -25°C and one minute shown in figure 15 clearly indicates that Royalene 5169 and polymer A have the lowest compression set at <70% because they both have lower ethylene content than the other EPDM polymers; while the other three EPDM polymers have considerably higher compression set at 80% or above. When the compression set is measured after relaxing at room temperature for 30 minutes, they all have comparable low temperature compression set properties in the range of 4% to 8%.

Figure 12 - crosslink density of Royalene 5169 and comparative EPDM polymers in low filler peroxide cure formulation

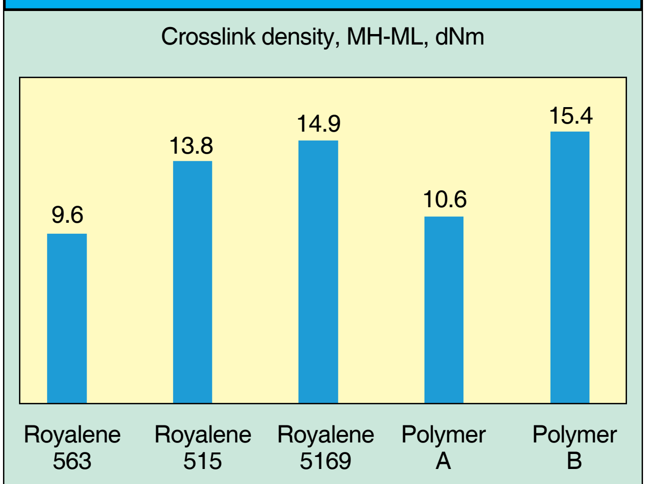


Figure 14 - original and heat aged elongation at break of Royalene 5169 and comparative EPDM polymers in low filler peroxide cure formulation

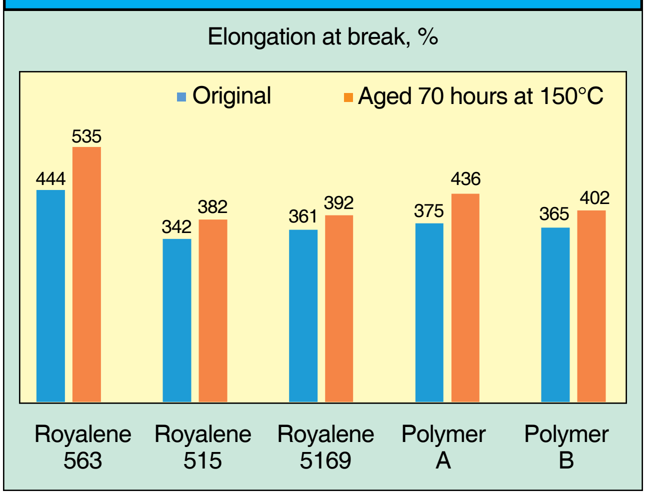
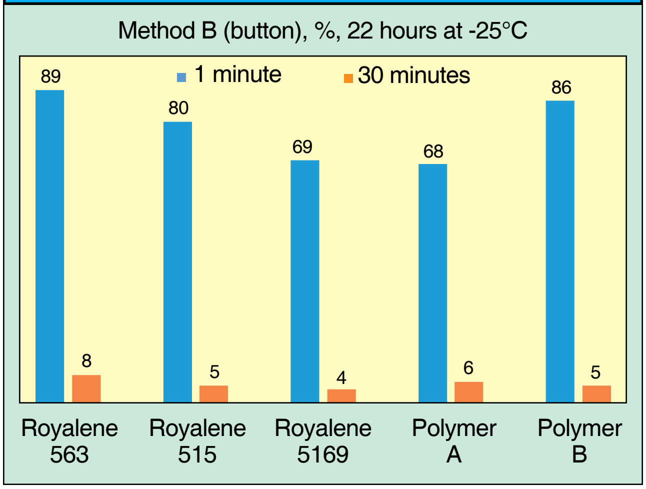


Figure 15 - compression set (22 hours at -25°C) of Royalene 5169 and comparative EPDM polymers in low filler peroxide cure formulation



Coolant hose (durometer 60 A, low sulfur cure)

EPDM polymer is the material of choice for automotive coolant hoses because of its excellent ozone crack resistance and heat aging characteristics, as well as its low temperature flexibility, chemical resistance and electrical properties, as mentioned before. In addition to these properties, coolant hoses require polymers with good processability during extrusion, and high green strength before curing in autoclaves with high pressure steam on mandrels. Therefore, it is a common practice to blend semi-crystalline EPDM polymer with amorphous EDPM polymer, and the semi-crystalline Royalene 539 and amorphous Royalene 563 have been successfully used in this application for decades.

Table 5 - coolant hose compound formulation with low sulfur cure

Formulation	*phr*
Polymer	100 + X
N-762 black	125
Calcium carbonate (GPR 325)	60
Paraffinic oil	70 - X
Zinc oxide	5
Stearic acid	1
Antioxidant 445	1
ZMTI	1.5
Sulfur	0.1
Masterbatch total	*363.6*
DTDM	2
ZDBC	2
DPTH	0.8
TMTD	0.8
Sulfur	0.4
Total	*364.5*

X: Oil content in the EPDM product

The addition of the ultra high molecular weight amorphous Royalene 5169 to the Royalene product slate provides another choice of EPDM polymer for this application, either by itself or in blends with other semi-crystalline EPDM polymers. In table 5, the Royalene 5169 is compared with some high Mooney amorphous EPDM polymers in a durometer 60 A coolant hose formulation with a low sulfur cure system. Table 6 shows the compound test results.

Figures 16-20 show the compound viscosity of these compounds, including the original and heat aged physical properties in a boiling 50/50 coolant/water solution. In figure 16, the EPDM polymers with ultra high molecular weight Royalene 5169 and polymer B have very high compound viscosity at 95 MU and 84 MU, respectively, due to their high molecular weights; while the other two EPDM polymers with lower molecular weight, Royalene 563 and polymer A, have lower compound Mooney at 50 and 55 MU, respectively. Figure 17 shows that Royalene 5169 and polymer B have the highest original tensile strength (>10 MPa), while Royalene 563 and polymer A have lower tensile strength at about 8.5 MPa because of their lower molecular weight. The compression set at -25°C and one minute shown in figure 18 clearly indicates that Royalene 5169 and polymer A have the lowest compression set at <50% because they both have lower ethylene content than the other two EPDM polymers, which have considerably higher compression set at 70% or above. The tensile strength change in a boiling 50/50 coolant/water solution for 168 hours and 1,000 hours is shown in

Figure 16 - compound viscosity of Royalene 5169 and comparative EPDM polymers in coolant hose low sulfur cure formulation

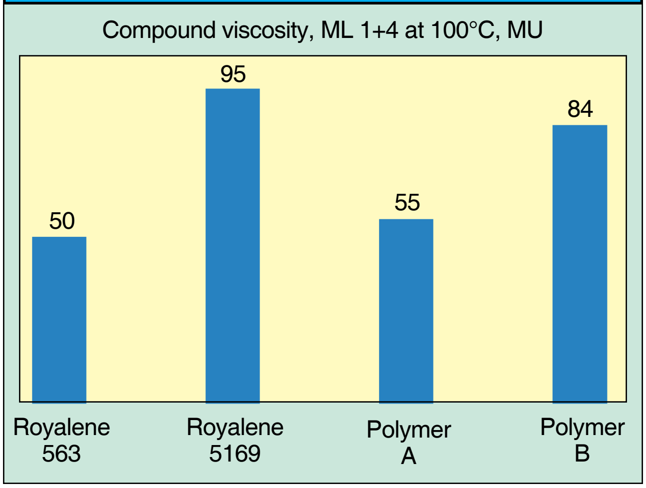


Figure 17 - tensile strength of Royalene 5169 and comparative EPDM compounds in coolant hose low sulfur cure formulation

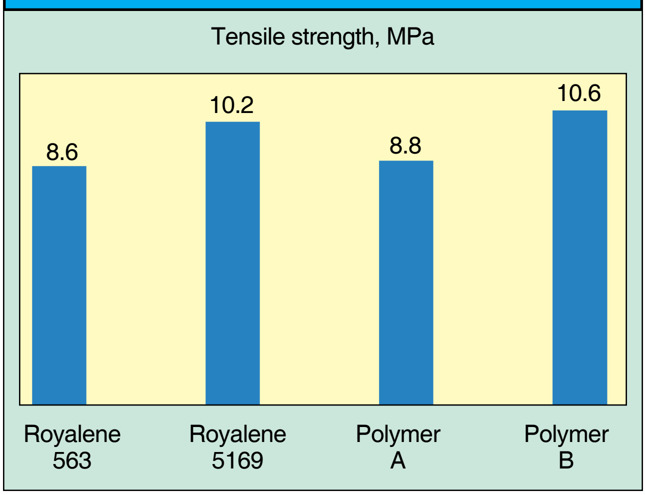


figure 19. The tensile strength of Royalene 5169 increased about 4% after both aging periods in a boiling coolant/water solution, while the tensile strength of other polymers decreased after 168 hours, then rose after 1,000 hours, probably caused by the hardening of the compounds. The volume swell data are shown in figure 20 in which the volume of Royalene 5169 and polymer B changed little (<0.2%) after 168 hours in the boiling coolant/water solution, while the volume of both Royalene 563 and poly-

Table 6 - coolant hose compound test results

Polymer	*Royalene 563*	*Royalene 5169*	*Polymer A*	*Polymer B*
Mooney viscosity, ML 1+4 at 100°C, MU	50	95	55	84
Mooney scorch, MS at 125°C				
t3, seconds	13.56	5.42	11.68	5.91
t35, seconds	-	-	-	-
Minimum viscosity at 125°C, MU	20	40	25	34
Rheometer at 165°C, 15 minutes				
ML, dNm	2.02	5.44	2.74	4.59
MH, dNm	17.74	23.58	21.04	23.24
ts2, minutes	2.29	1.42	1.92	1.61
t'c90, minutes	7.87	7.71	7.71	8.36
Cured properties (cure Tc 90 + 3 minutes at 165°C)				
Hardness, durometer A	60	64	62	64
100% modulus, MPa	1.9	3.4	2.3	3.1
Tensile strength at break, MPa	8.6	10.2	8.8	10.6
Elongation at break, %	540	300	433	349
Tear strength, die C, kN/m	24	20	20	22
Temperature retraction				
TR-10, °C	-41.4	-41.9	-47.0	-34.7
Physical properties, aged				
Heat aging in hot air oven (168 hours at 150° C)				
Hardness, durometer A	76	78	77	77
100% modulus, MPa	6.2	10.5	8.4	9.5
Tensile strength at break, MPa	10.9	11.8	11.1	11.6
Elongation at break, %	175	105	139	120
Tear strength, die C, kN/m	21	17	17	17
Compression set % (cure Tc 90 + 13 minutes at 170°C)				
Method B (button), deflection 25%; 22 hours, -25°C (1 minute)	70	48	44	76
Method B (button), deflection 25%; 22 hours, -25°C (30 minutes)	6	1	4	4
Method B (button), deflection 25%; 168 hours, 150°C	77	68	77	71
Immersion, 50/50 coolant/water, 168 hours at boiling point				
Hardness, durometer A	62	64	62	65
100% modulus, MPa	2.7	5.3	3.4	4.5
Tensile strength at break, MPa	9.0	12.3	9.9	11.8
Elongation at beak, %	395	191	289	226
Hardness, points change	-14	-14	-15	-13
Tensile, % change	-17	4	-11	2
Elongation, % change	126	81	107	88
Volume, % change	1.0	0.1	1.0	0.2
Weight, % change	2.1	1.6	1.8	1.6
Immersion, 50/50 coolant/water, 1,000 hours at boiling point				
Hardness, durometer A	63	66	64	65
100% modulus, MPa	3.3	6.3	4.4	5.7
Tensile strength at break, MPa	10.0	12.3	11.2	12.6
Elongation at break, %	289	173	229	200
Hardness, points change	-13	-12	-13	-12
Tensile, % change	-8	4	1	9
Elongation, % change	66	64	64	66
Volume, % change	2.6	3.1	2.7	3.4
Weight, % change	2.2	2.9	3.2	2.9

Figure 18 - compression set of Royalene 5169 and comparative EPDM compounds in coolant hose sulfur cure formulation

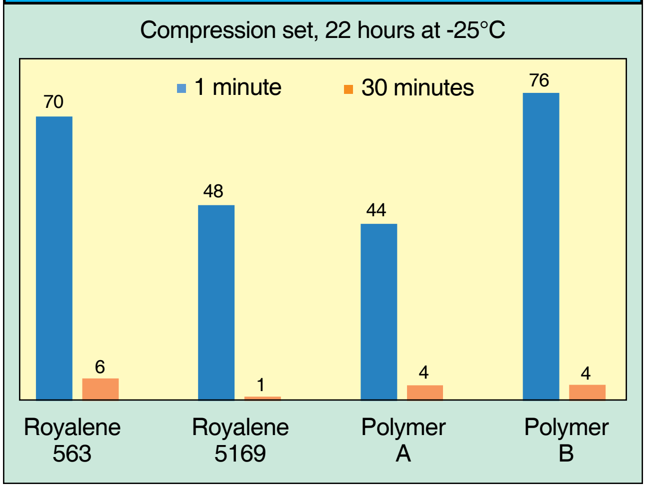
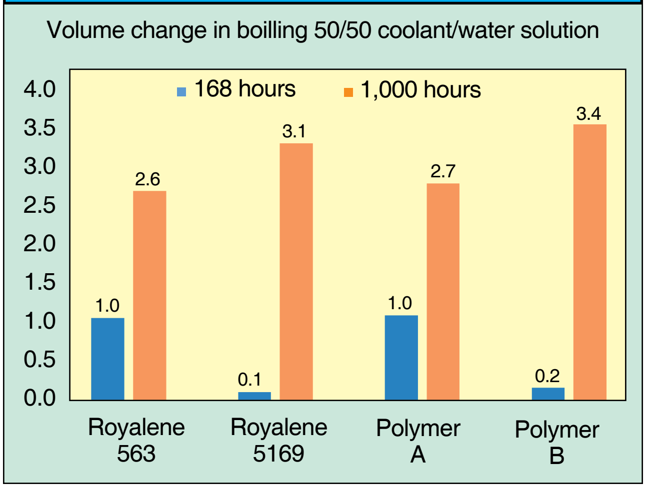


Figure 20 - volume change of Royalene 5169 and comparative EPDM compounds in coolant hose low sulfur cure formulation



mer A changed 1.0%. After aging in the boiling coolant/water solution for 1,000 hours, all the polymers had volume swell in the range of 2.6% to 3.4%; not significantly different.

Low hardness mechanical goods compound (durometer 45 A, sulfur cure)

In mechanical goods applications either, by molding or extrusion, the products sometimes need to have low hardness for applications such as grommets and architectural seals. The EPDM polymer used in these applications must have the ability to retain the high amount of oil in the compounds, while maintaining good processability and physical properties. Table 7 shows a typical durometer 45 A mechanical goods formulation in which an oil extended semi-crystalline Royalene 694 product (E/P: 70/30, ENB: 4.5 wt.%, Mooney: 50 ML 1+4 at 125°C, oil: 75 phr) commonly used for this application is included for comparison.

Figures 21-24 show the compound viscosity and cured physical properties of these compounds. In figure 21, only the EPDM polymers with ultra high molecular weight Royalene 694, Royalene 5169 and polymer B have reasonably high compound viscosity for processing, either for molding or extrusion. Figure 22 shows that all the polymers have comparable hardness and heat aging resistance. In figure 23, Royalene 694 has the highest original tensile strength (~12 MPa) because of its higher crystallinity in the polymer at room temperature, while Royalene 5169 and polymer B have comparable tensile strength at about 10 MPa, and Royalene 515 and polymer A have lower tensile strength because they have both lower ethylene content and molecular weight. The heat aged (70 hours at 125°C) tensile strength of these polymers follows the same trend. The compression set at -25°C and one minute shown in figure 24 clearly indicates that Royalene 5169 and Royalene

Figure 19 - tensile strength change of Royalene 5169 and comparative EPDM compounds in coolant hose low sulfur cure formulation

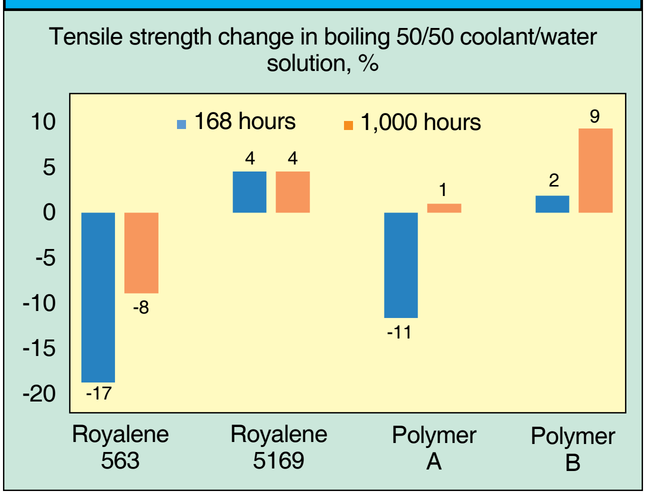


Figure 21 - compound viscosity of Royalene 5169 and comparative EPDM compounds in durometer 45 A mechanical goods sulfur cure formulation

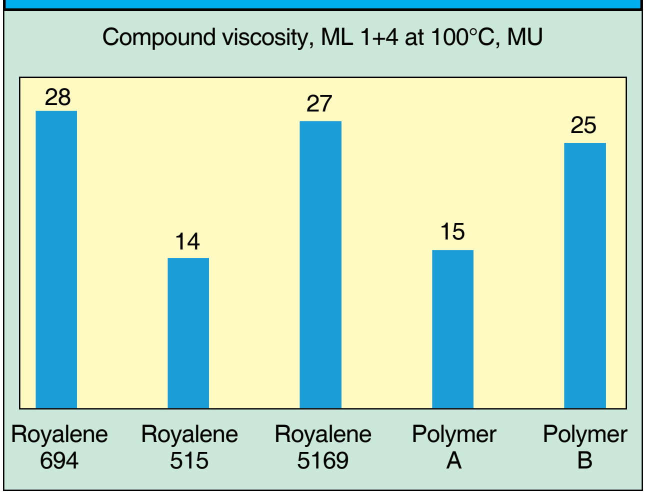


Table 7 - durometer 45 A mechanical goods formulation with sulfur cure

Formulation	*phr*
Polymer	100 + X
N347 black	100
Calcium oxide	15
Paraffinic oil	175 - X
Zinc oxide	20
Zinc stearate	1.5
Sulfur	0.1
Masterbatch total	*411.6*
MBTS	3
ZDBC	2.5
TMTD	0.8
DTDM	0.8
Sulfur	1.4
Total	*420.1*

X: Oil content in the EPDM product

515 have lower compression set at <45%, while the EPDM polymers A and B have higher compression set at about 55% because they have either lower molecular weight or higher ethylene content. The Royalene 694 has the highest compression set, as it is a semicrystalline EPDM polymer with high ethylene content. When the compression set was measured after the test buttons had relaxed at room temperature for 30 minutes, they all had comparable low temperature compression set properties at 10% or less.

Summary and conclusions

A wide variety of Royalene EPDM types are available, varying in Mooney viscosity, molecular structure, ethylene/propylene ratio and cure rate. The competitive landscape continually pushes EPDM users to be more cost competitive using the same types of polymers for specific applications, and thus demands the same of their suppliers. The market demanded an amorphous EPDM product which can be highly filled while maintaining its processability and low temperature compression set properties. A new experimental EPDM polymer, Royalene 5169, has been developed to satisfy these requirements. The key characteristics of the EPDM polymer are that it is an amorphous, medium diene product with ultra high molecular weight which maintains friability and is easy to mix. This EPDM was tested in several formulations, from highly extended compounds to very low hardness compounds. The compound test data show that in the highly extended formulations it maintained higher than 7 MPa tensile strength. It also had excellent tensile strength and low temperature compression set at high oil loading.

Figure 22 - hardness of Royalene 5169 and comparative EPDM compounds in durometer 45 A mechanical goods sulfur cure formulation

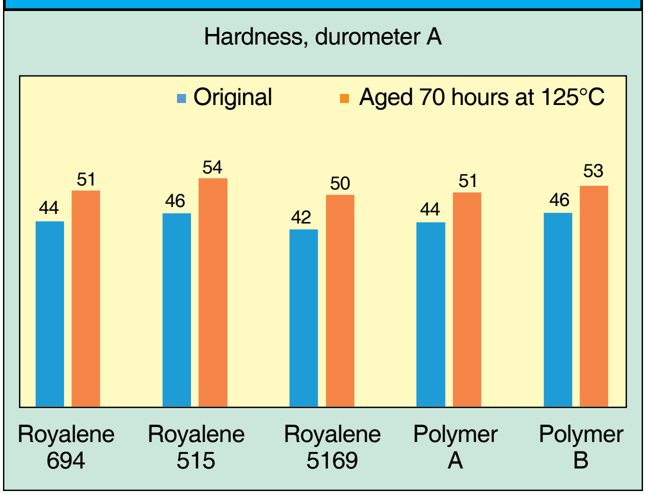


Figure 23 - tensile strength of Royalene 5169 and comparative EPDM compounds in durometer 45 A mechanical goods sulfur cure formulation

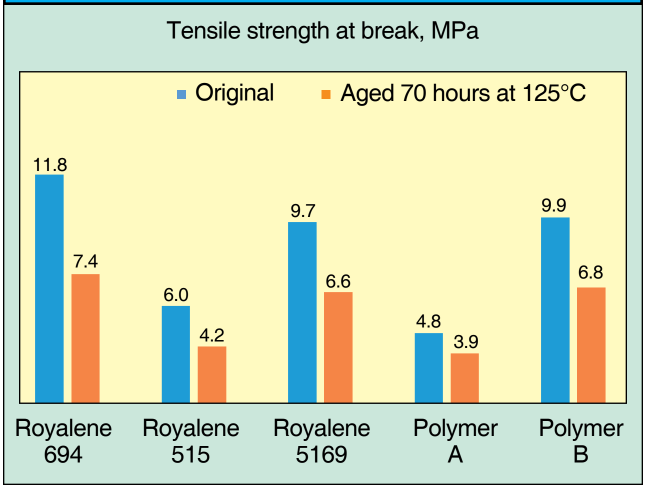
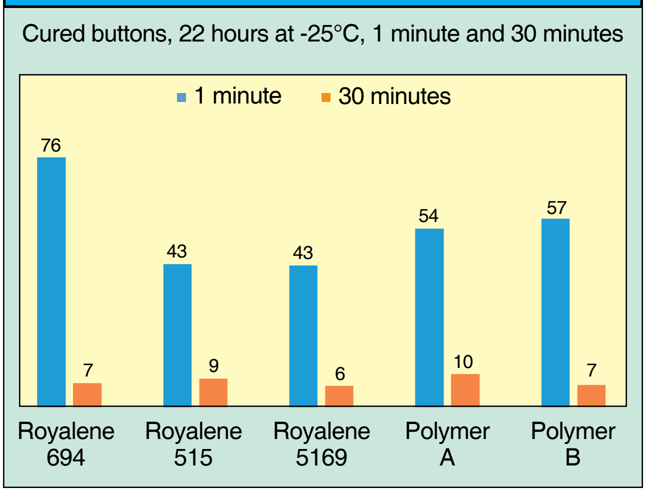


Figure 24 - compression set of Royalene 5169 and comparative EPDM compounds in durometer 45 A mechanical goods sulfur cure formulation



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