Isobutylene Elastomer Compound Optimization for Winter Tire Tread Applications[1]

Brominated Poly(isobutylene-co-para-methylstyrene) · dynamic properties · BIMS/BR/NR-blends · tire tread · wet traction handling properties · life time

Use of brominated isobutylene-co-para-methylstyrene (BIMS) improves lab dynamic properties predictive of wet and winter traction of formulations modeling winter passenger tire treads. Statistically designed blends of BIMS with cis-polybutadiene (BR) and natural rubber (NR) were studied in silane-coupled, silica-filled compounds. BIMS elastomer use requires adjustments to the curative system. By reducing sulfur and/or accelerator levels, compound cure and mechanical properties can be maintained.

Winter and wet traction and handling properties were measured for retreaded winter tires containing BIMS/NR/BR blends in the model winter tread. Performance was evaluated by independent tire testing labs for tires having three different levels of BIMS used in place of NR. Results were compared to tires with a solution-polymerized styrene-butadiene rubber/NR/BR model tread used as control, and to a commercial winter tire tread compound used as a reference. Vehicles having tires with BIMS winter treads showed reduced braking distances on an indoor ice surface, on a snowy glacier in Austria, on a wet asphalt surface, and on a dry surface compared to both the control and reference winter treads. The 20-phr BIMS tread had a 76,000-kilometer projected wear-out life based on road testing in the winter in the U.S.

Optimierung von Isobutylen-Compounds für PKW-Winterlaufflächen

Bromiertes Poly(isobutylen-co-p-methylstyrol) · dynamische Eigenschaften · BIMS/BR/NR-Verschnitte · Laufflächen · Nassrutschen · Fahreigenschaften · Lebensdauer

Der Einsatz von bromiertem poly(isobtylen-co-p-methylstyrol) (BIMS) verbessert die Voraussagbarkeit des Rutschverhaltens bei Nässe und Schnee von PKW-Modelllaufflächen. Nach einem statistischen Versuchsplan erstellte Verschnitte bestehend aus BIMS und BR bzw. NR wurden in Kieselsäure/Silan Compounds untersucht. Der Einsatz von BIMS erfordert die Anpassung des Vernetzungssystems. Eine Verminderung der Schwefel und/oder Beschleunigerkonzentration führt nicht zu Einbußen des mechanischen Eigenschaftsbildes. Wintereigenschaften, Nassrutschen und das Fahrverhalten wurden an runderneuerten Verschnitten aus BIMS/NR/BR in Modellaufflächen untersucht. Die Eigenschaften wurden in unabhängigen Reifenprüflaboratorien untersucht. Die Ergebnisse wurden mit Modellaufflächen aus L-SBR/NR/BR und kommerziellen Winterreifenlaufflächen verglichen. Fahrzeuge mit BIMS-Winterreifenlaufflächen zeigen zu beiden Bezugssystemen einen verminderten Bremsweg auf einer indoor Eisfläche, auf verschneiten Gletscherflächen, nassem Asphalt und auf trockenen Oberflächen. Die Lebensdauer von Laufflächen mit 20 phr BIMS wird auf der Grundlage von Winterstraßentests auf 76 000 km geschätzt.

Brominated isobutylene-co-para-methylstyrene (BIMS), see *Fig. 1*, is commercially known as the Exxpro^{TM} elastomer. It can contain between 2 and 8 moles of para-methylstyrene monomer per 100 moles of isobutylene monomer, and can have bromine levels ranging from 20–50 % of the para-methylstyrene content. Rogers and Waddell [1] have reviewed laboratory studies and tire evaluations of the BIMS polymer in a variety of tire compounds including inner liner, black sidewall and tread formulations.

Mroczkowski [2] used blends of BIIR, star-branched-BIIR and BIMS in carbon black/silica-filled SBR / BR compounds to increase tangent delta values at – 30 °C to + 10 °C and decrease tangent delta values at temperatures above 30 °C compared to a carbon black-filled BR/NR/SBR tire tread; laboratory abrasion resistance was comparable. Costemalle, Hous and McElrath [3] used BIIR or BIMS to increase the tangent delta at 0 °C for carbon black-filled emulsion and solution SBR/BR compounds. Zanzig, Sandstrom, Verthe and Crawford [4] used BIMS in blends with IBR, BR, and/or SBR in silane-coupled silica-filled compounds to increase tangent delta at 0 °C. Kadomaru and Nakada [5] increased tangent delta at 0 °C using 15-phr BIMS with SBR. Rogers [6] used BIMS and silane-coupled silica in BR/SBR compounds to increase tangent delta at 0 °C and decrease tangent delta at 60 °C in laboratory tests, with only slight reductions in tread-wear based on tests using sectional retreaded tires. Hojo [7] used a hydrazide compound with BIMS to lower the heat generation and improve the wet gripping property of carbon black/silane-coupled silica-filled NR. Muraoka, Minigawa and Yagi [8, 9] studied silane-coupled silica-filled BIIR/SBR and BIMS/SBR treads showing improved wet skid performance with comparable laboratory abrasion.

Tire testing has shown the use of BIMS to be a traction-improving polymer. Matsui and Ohhashi [10] used BIMS to improve tire snow and ice and wet performances; wear resistance was maintained. Hara and Muraoka [11] used BIMS to increase the wet grip performance and the "feeling of driver" in tire tests. Minigawa, Muraoka and Kakumaru [12] reported an increase in tire wet grip, reduction in rolling resistance, and maintained abrasion resistance using BIMS with NR/SBR and with NR. Nakadera, Wada, Mine and Sugiyama [13] studied silane-coupled silica-filled BIIR/SBR and BIMS/SBR treads, and showed improved wet skid performance on kart tires using 3,4-polyisoprene.

We [14–20] previously used BIMS in place of sSBR in an all-season passenger tire tread modeled on a silica-filled BR/sSBR passenger tire tread [21], and with NR / BR in a model winter passenger tire tread. Laboratory dynamic properties predicted [22] potentially improved wet/winter traction and rolling resistance. Sectional retread tire tests of both the model all-season

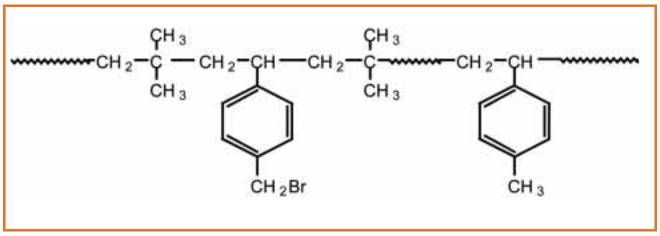

Fig. 1. Structure of brominated isobutylene-co-para-methylstyrene, BIMS

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and winter passenger tire treads established the road wear potential of BIMS approaching 76,000 kilometers (48,000 miles) [19, 20]. This report focuses on the winter and wet traction and handling performances of sectional retread winter tires with BIMS elastomer treads.

Experimental

Materials. Brominated isobutylene-co-para-methylstyrene (BIMS) is Exxpro$^{\text{TM}}$ 3745 from ExxonMobil Chemical containing 7.5 weight-% para-methylstyrene comonomer and having 1.2 mole-% brominated para-methylstyrene. cis-1,4-Polybutadiene is Buna CB23 (96 % cis) from Bayer. Natural rubber is SMR 20. Solution-polymerized styrene-butadiene is NS116 (20 % bound styrene) from Nippon Zeon. All other compound ingredients were obtained from commercial sources. See the BIMS (#1 – #3) and sSBR control (#4) formulations in *Tab. 1*. Compounds were mixed in a three-step mixing sequence using a 45-liter Krupp internal mixer with PES-5 interlocking rotors at their facility in Germany. The mixing procedure is also shown in Tab. 1. A two-roll mill was used to sheet out stocks after each mixing step. A fifth compound was used as a reference for all tire testing: a winter tread compound used on commercial passenger winter tires in Europe.

Test Methods. When possible, ASTM tests were used to determine the cure and cured compound physical properties. Cure properties were measured using a MDR 2000 (0.5 arc) at 160 °C. Test specimens were cured at 160 °C for a time corresponding to Tc90 + 2 minutes for mold lag. Stress/strain properties (tensile strength, modulus, elongation at break, energy to break) were measured at room temperature using an Instron 4202. Shore A hardness was measured at room temperature using a Zwick Duromatic. Abrasion loss was determined at room temperature by weight difference by using an APH-40 Abrasion Tester with rotating sample holder (5 N counter balance) and rotating drum. Weight losses were indexed to that of the standard DIN compound with lower weight losses indicative of a higher abrasion resistance index. Temperature-dependent (– 80 °C to 60 °C) dynamic properties (G^*, G', G'' and tangent delta) were measured using a Rheometrics ARES mechanical spectrometer at 1 Hz or 10 Hz and 2 % strain using a rectangular torsion sample geometry cut from a standard cured tensile sheet. Temperature-dependent (– 90 °C to 60 °C) high-frequency acoustic measurements [23, 24] were also performed at Sid Richardson Carbon Company using a frequency of 1 MHz and ethanol as the fluid medium.

Tire Retreading. Five sets of retreaded tires were prepared by Unigom NV, Belgium to evaluate the effects of polymer adjustments to the BIMS model winter tire tread formulation. New Michelin Alpine P195/65R15T winter passenger tires from the same production lot were buffed to remove the tread and sidewall. A layer of commercially available sidewall and cushion gum compounds were placed over the exposed carcass and stitched automatically. The tread sections were plied to the cushion gum and stitched automatically. Tires were cured from 24 to 40 minutes at 150 °C in a mold to afford the highly

Tab. 1. Model Winter Tire Tread Compounds

Ingredients	Time (sec)	#1 (phr)	#2 (phr)	#3 (phr)	#4 (phr)
1st Step: Fill factor = 65 %, Rotor speed = 65 rpm, TCU = 40 °C, Ram pressure = 6 bars					
BIMS, Exxpro$^{\text{TM}}$ 3745	0	20	30	40	
BR, Buna CB 23	0	40	40	40	40
NR, SMR 20	0	40	30	20	30
sSBR, NS116	0				30
Silica, Zeosil 1165MP	30	60	60	60	60
Silane, X50S	30	10.2	9.4	8.6	12
Silica, Zeosil 1165MP	120	15	15	15	15
Processing Oil, Mobilsol 30	120	30	30	30	30
Dump time (160 °C maximum)	375				
2nd Step: Fill factor = 65 %, Rotor speed = 50 rpm, TCU = 40 °C, Ram pressure = 6 bars					
Accelerator, DPG		2	2	2	2
Stearic acid		1	1	1	1
Antiozonant, Santoflex 6PPD		1.5	1.5	1.5	1.5
Antioxidant, Agerite Resin D		1	1	1	1
Dump time (137 °C maximum)	120				
3rd Step: Fill factor = 65 %, Rotor speed = 25 rpm, TCU = maximum cooling, Ram pressure = 6 bars					
Activator, Zinc oxide		2	2	2	2
Curative, Sulfur		1	1	0.8	1
Accelerator, Santocure TBBS		1.5	1.5	1.2	1.5
Accelerator, Thiate U				0.25	
Dump time (110 °C maximum)	110				

Fig. 2. MS Botrange winter passenger retread pattern

siped winter tire tread pattern of the MS Botrange, see *Fig. 2*.

In addition, retreaded tires each comprising sections of the five tread compounds (three BIMS, sSBR control, and commercial reference) were also prepared for tread wear testing. Each compound section was randomized on the four tires such that no two compounds had the same relative location on any tire.

Tire Testing. Retreaded winter tires were tested by TÜV Automotive GmbH, Germany. Ice testing was done at an indoor rink in Kaufbeuren, Germany. The ice temperature was – 4.2 °C to – 2 °C and the air temperature ranged from 3 °C to 6.8 °C. Tires were equilibrated to the ice temperature, mounted on a Mercedes C200 Kompressor with ABS brakes and approximately 900-kg load on each axle, inflated to 2.5 bar, and tested for braking and cornering acceleration. Braking distance was determined by having the professional drivers applying full, constant pressure to the brake pedal and using a Doppler radar sensor to measure the distance to slow the car from 20 km/h to 5 km/h. Cornering acceleration was determined by measuring the lap time around an 8-meter radius circle with the vehicle being driven as fast as was safely possible.

TÜV Automotive testing of the Alpine winter properties of acceleration, braking and handling was performed on the glacier of Solden, Austria. The distance needed to accelerate the Mercedes C200 Kompressor from 20 km/h to 35 km/h was measured. The distance to slow down the vehicle from 40 km/h to 5 km/h was measured. The snow and air temperatures during these tests ranged from – 10 °C to – 4 °C, respectively. Snow handling is considered a subjective test and was carried out by two professional drivers who judged traction, steering, cornering, accelerating and braking behaviors on an uphill and downhill sinuous track while the vehicle was being driven at both low and high speeds. The snow and air temperatures of this test were similar, ranging from – 5 °C to – 3 °C.

Wet braking, handling and longitudinal aquaplaning were determined by TÜV Automotive using a Mercedes C200 Kompressor at the MIRA Wet Handling Circuit in Warwickshire, U.K. The distance was measured to slow down the vehicle from 80 km/h to 15 km/h while traveling in an artificially rained asphalt track having a water depth of 2 mm. The water and air temperatures ranged from 13 °C to 18 °C. Wet handling is also considered to be a subjective test and was again carried out by two professional drivers who judged lateral tracking, steering, cornering, accelerating and braking behaviors on a sinuous artificially rained track. Longitudinal aquaplaning was measured on a straight course as the speed of the vehicle at which time the slip on the wet surface (8-mm of water) was > 15 % than that on a dry surface. Lateral aquaplaning was determined on the Semperit Circuit in Traiskirchen, Austria using a 200-meter diameter circular course with the Mercedes C200 Kompressor driven at a set steering angle through a 20-meter section having a 5-mm water depth. The highest speed that the vehicle attains in the wet curve before losing grip was recorded, and the professional driver noted the ability to regain steering control of the vehicle. The water and air temperature ranged from 13 °C to 18 °C.

Ice and snow properties were also measured under the severe Nordic winter conditions at Test World in Ivalo, Finland, where both the daytime ice and the air temperatures were – 10 °C. Tires were

Tab. 2. Model Winter Tread Cure and Cured Properties

Properties	#1	#2	#3	#4
MDR 2000E @ 160 °C				
MH-ML (dN.m)	13.34	10.43	7.92	15.55
Ts2 (min)	2.42	3.21	3.3	2.16
Tc50 (min)	3.77	4.68	4.96	3.63
Tc90 (min)	5.13	6.45	8.49	5.10
Hardness, Shore A @ 23 °C	56	56	54	57
20 % Modulus (MPa)	0.78	0.72	0.55	0.79
100 % Modulus (MPa)	1.81	1.76	1.52	1.93
300 % Modulus (MPa)	8.86	9.20	7.34	9.16
Tensile at Break (MPa)	16.69	14.71	12.21	21.06
Elongation at Break (%)	484	427	430	555
DIN Abrasion Index	153	134	117	162
ARES Rheometrics @ 1 Hz				
G* @ 60 °C (MPa)	3.80	4.46	4.24	5.24
Tan Delta @ 60 °C	0.05	0.05	0.04	0.09
G′ @ 30 °C (MPa)	4.00	4.73	4.36	5.71
Tan Delta @ 30 °C	0.07	0.08	0.06	0.13
G″ @ 0 °C (MPa)	0.73	0.80	0.71	1.51
Tan Delta @ 0 °C	0.16	0.17	0.15	0.19
G* @ – 30 °C (MPa)	13.03	13.84	15.16	15.90
Tan Delta @ – 30 °C	0.55	0.46	0.62	0.39
High Frequency Acoustic Testing				
Longitudinal Coefficient @ 25 °C	0.068	0.083	0.102	0.053
Tire Testing				
Hardness, Shore A @ 23 °C	60	58	56	61
Projected Wear-out[20] (1000 kilometers)	76.9	62.8	57.3	78.2

mounted on a Saab 9-5 2.0t sedan or Saab 9-3 2.0t wagon with ABS brakes, and tested for braking and cornering acceleration. The ice surface was conditioned with a machine prior to individual tests. Braking distance was determined by having the professional driver apply full, constant pressure to the brake pedal and using a Doppler radar sensor to measure the distance it takes to slow the car from 20 km/h to 5 km/h. Cornering acceleration was determined by measuring the lap time around a 25-meter radius circle. Acceleration, braking and handling in frozen snow was performed. The snow was compacted to form a hard surface prior to each test. The distances needed for the professional driver to accelerate the car from 10 km/h to 35 km/h, and to slow down the vehicle from 35 km/h to 5 km/h were measured. Snow handling is a subjective test carried out by two professional drivers judging traction, steering, cornering, accelerating and braking behaviors of the vehicle while driving as fast as possible on the track. The time to complete three successive laps is one consideration.

Dry braking was measured at the TÜV proving ground in Jesenwang, Germany. The distance to slow down an Audi A4 1.9 TDI from 80 km/h to 15 km/h was measured. The surface temperature ranged from 15 °C to 18 °C, and the air temperature ranged from 10 °C to 13 °C. Dry handling was tested at the Nurburgring at Nordschleife, Germany. It is also considered a subjective test, and was carried out by two professional drivers judging lateral tracking, steering behavior and driving stability on the 21-km sinuous track. The surface temperature ranged from 22 °C to 39 °C, and the air temperature ranged from 14 °C to 20 °C.

A bock tear-out test was also performed at the Nurburgring by running 10 laps in an Audi A4 1.9 TDI at a speed of about 120 km/h. Lap times ranged from 10.25 to 10.5 minutes. Tires were inspected after each lap. The surface temperature ranged from 12 °C to 30 °C, and the air temperature ranged from 9 °C to 16 °C.

High-speed treadwear was performed at the Nardo Proving Ground "Prototipo" in Italy. Identical Audi A4 1.9 TDI front wheel drive passenger cars were driven at the same time in procession at an average speed of 178 km/h on a round track having a radius of 2 km and length of 12.57 km until 15,000 km was driven. The track had an almost horizontal longitudinal profile, with the ring sloping towards the center of the track to compensate for the centrifugal force and permitting speeds between 100 km/h and 240 km/h without the driver having to change direction significantly. Drivers and tires were interchanged between the cars at regular intervals to randomize any differences between the performance of the car or driver. Driving direction was alternated daily. The average air temperature during the testing period varied between 8 °C and 16 °C; on three sunny days the temperature rose to 20 °C. The road was mostly dry, only for approximately 1000 km the surface was wet or moist. Front axle load of 900 kg, rear axle load of 610 kg. Every 1500 km, the tread pattern depth was measured at four positions along the tire circumference in 5–6 groves, but the tires were not rotated. After 15,000 km of testing, the projected wear to the wear bar indicator was calculated.

Tread wear was also tested by the Nevada Automotive Test Center near Carson City, NV in the U.S. on the five-sectional retreaded tires. Each tire had equal sections of the 20-phr, 30-phr and 40-phr BIMS, the sSBR control and the commercial reference compounds. In addition, each compound was randomized such that no two compounds had the same relative location on each tire. Weight was added to the GMC Sonoma to place a 508-kg load on each tire at 2.4-bar pressure, and the vehicle ran a test circuit of primary paved road surfaces that included 15 % hill and curve mountain ascent and descent. Speeds ranged from 24 kilometers per hour to 112 kilometers per hour (Interstate highway), averaging 95.4 km/h. Air temperatures ranged from −12 °C to +13 °C. The test sections were measured every 2560 kilometers at three designated points on each section for tread depth until 21,900 kilometers was reached. Tire position was rotated at each tread depth measurement period.

Results and Discussion

BIMS Model Winter Tire Tread Compounds. An extreme-vertices mixture designed experiment was used to balance the BIMS, NR and BR polymer ratios in order to maximize the laboratory DIN abrasion resistance while maintaining the potential for improving traction properties based upon measured tangent delta values [16, 19] from Rheometrics ARES testing. The highest laboratory abrasion resistance values were obtained with the highest BR values studied (40 phr), which is consistent with the traditional use of high cis-BR to improve tire tread wear. In the present tire traction and handling evaluations, BIMS was thus used in place of NR, while maintaining BR at 40 phr, see *Tab. 2*. The reduction in NR levels and corresponding increases in BIMS concentrations shifted the peak of the tangent delta curve to higher temperatures and also broadens this curve, see *Fig. 3* [19, 20]. This higher, broader tangent delta peak shows the potential for increased wet/winter traction properties using higher levels of the BIMS elastomer. The high-frequency acoustic temperature sweeps also show that the maximum of the longitudinal coefficient values is shifted to higher temperatures when using tread compounds having increasing levels of BIMS, see *Fig. 4* [19].

Tire Testing. Vehicle response under Nordic winter conditions (ice and compacted frozen snow) were measured under the severe service winter conditions in Ivalo, Finland. When the vehicle response equipped with tires having model BIMS tread compounds was normalized to that of the commercial winter tread compound used as a reference, results varied. Tires with the 30-phr and 40-phr BIMS treads afforded generally lower braking, traction and handling performance on both the frozen snow and ice. On ice, the tires with the 20-phr BIMS tread gave somewhat better performance than did the reference. *Tab. 3* shows relative test results obtained by indexing values to that of the reference, which was assigned a value of 100. Indexed values greater than +/−2 % are thought to be a significant effect.

Braking distances were also measured at an indoor ice rink. Under these milder simulated winter conditions, the vehicle braking distances of tires with the three BIMS model tread compounds were up to 15 % shorter than that of the commercial reference and sSBR control compounds as indicated by the higher index values, see Tab. 3. The laptimes of vehicles with tires having the BIMS model treads were slightly shorter than that equipped with the commercial reference compound. Due to the 8 °C to 10 °C higher temperatures of the air than that of the ice surface inside the indoor rink, a water layer is presumed to be present on the ice surface, better simulating Alpine winter conditions of cold wet traction conditions than of frozen ice.

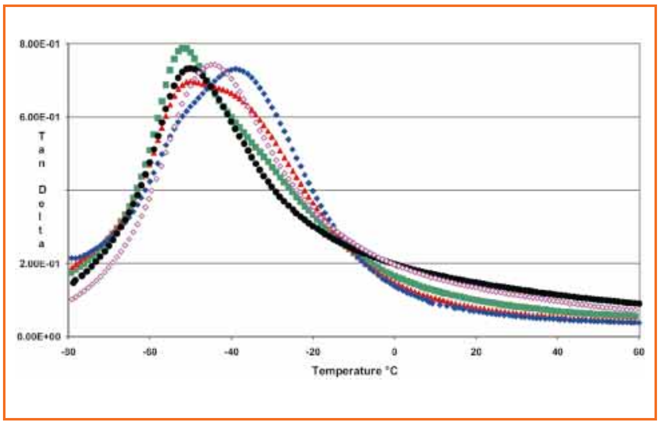

Fig. 3. Tangent delta (1 Hz, 2 % strain) of sSBR control NS116 (black circle) and 20 phr BIMS (green square), 30 phr BIMS (red triangle), 40 phr BIMS (blue diamond), and commercial reference (open purple diamond) winter tread compounds

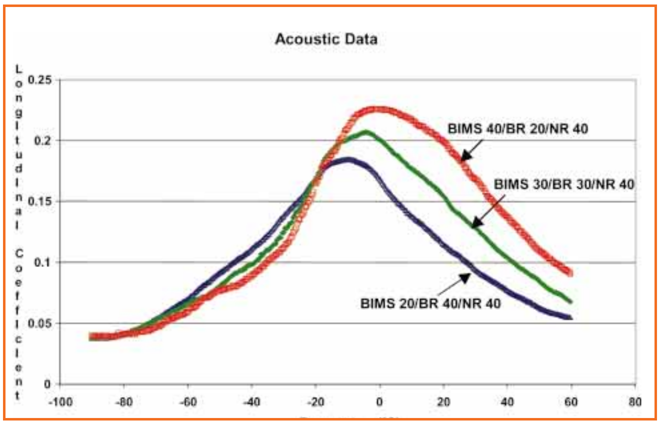

Fig. 4. Temperature-dependent high-frequency acoustic curves for BIMS model winter tread Compounds #1–#3

Tab. 3. Relative Winter Tread Braking, Traction and Handling Properties

Property	#1	#2	#3	#4	#5
	20-phr BIMS	30-phr BIMS	40-phr BIMS	30-phr sSBR	Commercial Reference
Nordic ice					
– Braking distance	104	101	98	107	100
– Laptime	101	98	98	105	100
Nordic snow					
– Braking distance	99	97	97	102	100
– Traction	100	96	95	102	100
– Handling	98	97	96	100	100
Indoor ice					
– Braking distance	113	115	105	96	100
– Laptime	103	102	106	107	100
Alpine snow					
– Braking distance	108	115	108	107	100
– Traction	94	98	99	93	100
– Handling	102	109	102	126	100
Wet					
- Braking distance	102	103	107		100
- Aquaplaning		103	102	100	100
- Handling			120		100
Dry					
- Braking		107	104		100
- Handling					100
High-speed wear					
- Projected wearout, front	197	191	132		100
- Projected wearout, rear	126	107	112		100
Wear					
- Projected wearout	124	103		126	100

Testing of vehicle acceleration, braking and handling properties on the snowy glacier in Solden, Austria showed that the braking distances were also shorter for tires with BIMS tread compounds compared to the reference. However, the measured tire traction for the three BIMS model treads was poorer than that of the reference, although comparable to or better than that of the sSBR control compound. The subjective snow-handling test indicated that the tires with BIMS model treads were acceptable, see Tab. 3.

Wet braking and handling properties of tires with the three BIMS model treads were slightly higher than that of the reference, and measurably higher than that of the sSBR control, see Tab. 3. Longitudinal aquaplaning was comparable. Testing of lateral aquaplaning showed that compared to the three BIMS model tread compounds, the vehicle with the sSBR control tires attained a higher speed before skidding, but the driver was not able to safely steer the vehicle out of the skid. Alternatively, drivers of the vehicles equipped with tires having the three BIMS model tread compounds could easily regain control of the vehicle and continue on the test run. Vehicles equipped with tires having the commercial reference tread compound afforded the best performance of speed and steering characteristics.

Braking of vehicles with tires having the 30-phr and 40-phr BIMS model treads was higher than that of tires with either the reference or control compounds on dry surfaces, see Tab. 3. However, vehicle handling was notably lower for all three tire sets with BIMS model treads compared to the reference or control compounds. However, this latter test was performed when the road surface temperature ranged from 22 °C to 39 °C, and is too hot a temperature to be representative of winter conditions.

Testing of tire tread wear of vehicles travelling at high speeds (average of 178 km/h) showed that the BIMS model compounds performed satisfactorily since projected

Tab. 4. Model Winter Tread Dynamic Properties

ARES Rheometrics @ 10 hz	#1	#2	#3	#4
G* @ 60 °C (MPa)	3.53	3.63	3.35	4.59
Tan Delta @ 60 °C	0.09	0.08	0.08	0.12
G′ @ 30 °C (MPa)	4.08	3.98	3.74	5.41
Tan Delta @ 30 °C	0.14	0.12	0.12	0.16
G″ @ 0 °C (MPa)	1.66	1.84	1.97	1.83
Tan Delta @ 8 °C	0.29	0.33	0.37	0.25
G* @ – 30 °C (MPa)	20.51	28.01	40.63	22.61
Tan Delta @ – 30 °C	0.64	0.73	0.80	0.65

wear out values were higher than that expected for tires having the commercial tread, see *Fig. 5*. However, some chip/chunking was observed under these test conditions. Tires with 40-phr BIMS treads showed the most severe condition, with little observed for the 20-phr BIMS compound. No chipping/chunking was observed for tires having either the 30-phr and 40-phr BIMS model treads during the block tear-out testing consisting of 10 laps at an average speed of 120 km/h when tested at the Nurburgring. Thus, chip/chunking is not thought to be a wear issue.

Tread wear at moderate vehicle speeds was determined using tires having 5-section retreads. The 20-phr BIMS tread sections had a projected wear-out of 76,900 km and the 40-phr BIMS sections had a projected wear-out of 57,000 km, see Tab. 2 [20]. Thus, the BIMS elastomer shows the wear resistance potential (50,000 km) to complete the two seasons required to be a viable passenger tire winter tread.

Correlation of Tire Performance to Laboratory Properties. The poorer Nordic winter test results obtained for the vehicles equipped with tires having the 30-phr and 40-phr BIMS model treads were not as expected. Based upon the higher tangent delta values measured at – 30 °C (see *Fig. 6*) or the lower G* values measured at – 30 °C (see Tab. 2), it was predicted that these BIMS model tread compounds could have performed better in these winter tire tests. Since previous laboratory temperature-dependent dynamic properties of isobutylene-based compounds showed the dependence upon testing frequency [18], compounds were re-tested using a frequency of 10 Hz, see *Fig. 7* and *Tab. 4*. These results show that the G* values at – 30 °C are higher for the 30-phr and 40-phr BIMS model treads than for the sSBR control compound, predicting the poorer winter properties [22, 25], which was observed.

Similarly, it was not expected that vehicles with tires having the BIMS model tread compounds would have improved performance on wet asphalt surfaces based upon predictions from tangent delta values measured at 0 °C, see Fig. 6 and Tab. 2. However, using dynamic properties measured at a frequency of 10 Hz, see Fig. 7 and Tab. 4, all three BIMS model tread compounds have higher tangent delta values at 0 °C, thus correctly predicting the improved wet traction properties observed. This is also in agreement with predictions based upon high frequency acoustic testing [23, 24] whereby increasing the BIMS level both increases and shifts the maximum value of the longitudinal coefficient to higher temperatures. Since traction and braking are high-frequency phenomena [26–28], the BIMS elastomer is expected to better serve as a wet traction polymer since, as is also the case for all isobutylene-based elastomers, there are two transitions [29]. The lower temperature transition at ca. – 55 °C is observed as a

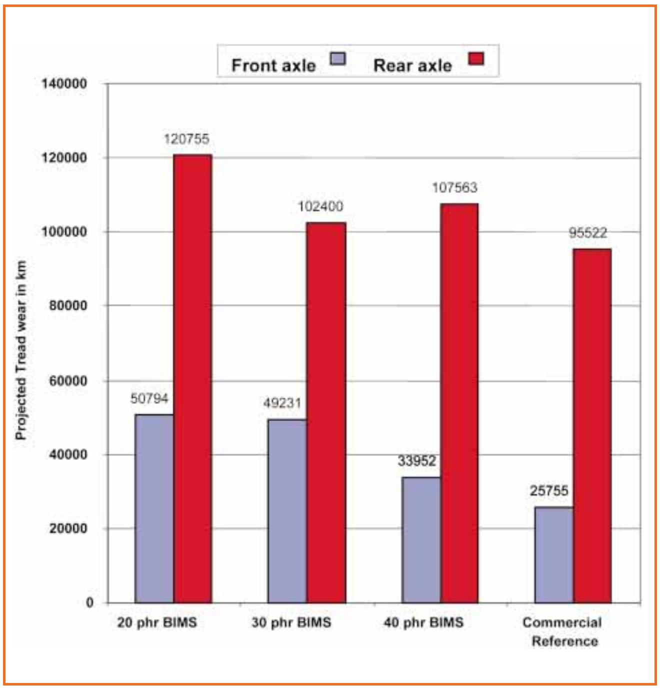

Fig. 5. High-speed (178 km/h) projected tread wearout

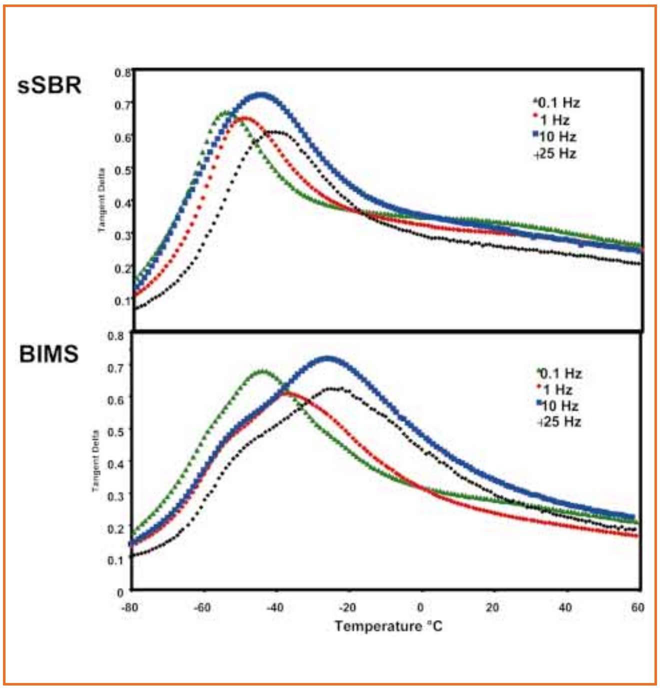

Fig. 6. Tangent delta versus temperature for sSBR/BR/NR (40/40/20) and BIMS/BR/NR (40/40/20) compounds tested at 0.1 Hz (triangle), @ 1 Hz (diamond), @ 10 Hz (square), and @ 25 Hz (plus sign)

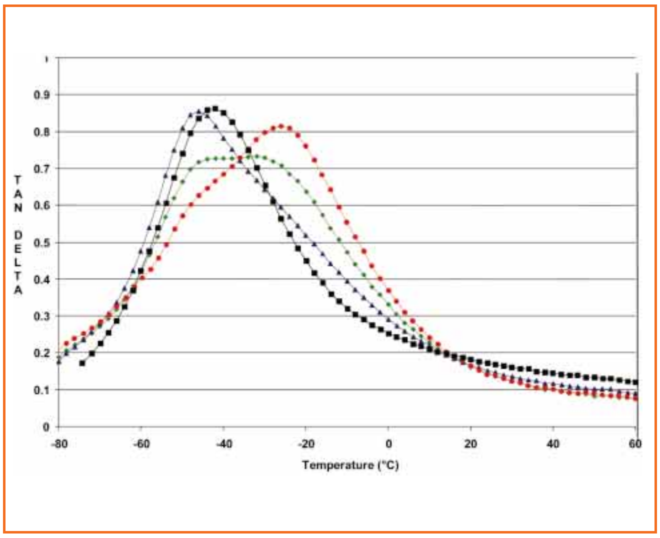

Fig. 7. Tangent delta (10 Hz, 2 strain) of sSBR control NS116 (black square) and 20 phr BIMS (blue triangle), 30 phr BIMS (green diamond) and 40 phr BIMS (red circle) model winter tread compounds

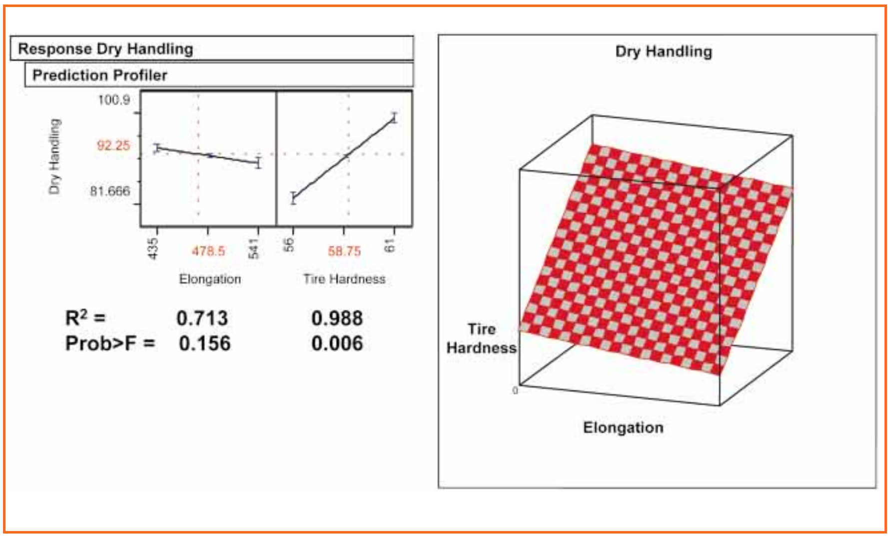

Fig. 8. SAS JMP statistical analysis of tire dry handling test data to compound properties

shoulder on the tangent delta peak for the BIMS compounds (Fig. 6) and is the Tg resulting from normal segmental motions, the movement of the Kuhn segment. The second higher-temperature transition above this Tg relaxation is most relevant to tire traction properties. This transition is due to local torsional motions along the polymer chain (sub-Rouse motion). In general, these segmental motions are coupled. However, isobutylene-based elastomers are unique in having weak intermolecular chain interactions due to its compact, symmetrical repeat unit containing the geminal dimethyl group. Segmental relaxation in polyisobutylene is so fast [29] that it is decoupled from the local torsional motion, which occurs subsequent to segmental relaxation (Tg). In addition, isobutylene-based elastomers are not thermorheologically simple, meaning that time-temperature superposition does not work.

The poorer dry handling performance observed for the BIMS model tread compounds compared to the commercial reference or the sSBR control tread was statistically analyzed by correlating the dry performance to all individual tread compound physical and dynamic properties. The primary variable identified as impacting dry handling performance was the Shore A hardness values measured on the retreaded tires, see Tab. 2. Regression analysis shows a very high correlation coefficient ($r^2 = 0.988$) and confidence level ($\text{Prob} > F = 0.006$) whereby the higher hardness compound affords the highest dry handling test ratings. Elongation to break ($r^2 = 0.713$) was the next most important variable identified; however, its impact upon dry handling is not significant ($\text{Prob} > F = 0.156$), see *Fig. 8*.

Finally, there is a significant correlation ($R^2 = 0.97$) with high confidence ($\text{Prob}>F = 0.0022$) between laboratory DIN abrasion index values and projected wear out, see *Fig. 9*. This may simply be a result of having all five tread sections being present in random order on all four tires, and each tire was rotated through each position on the vehicle twice.

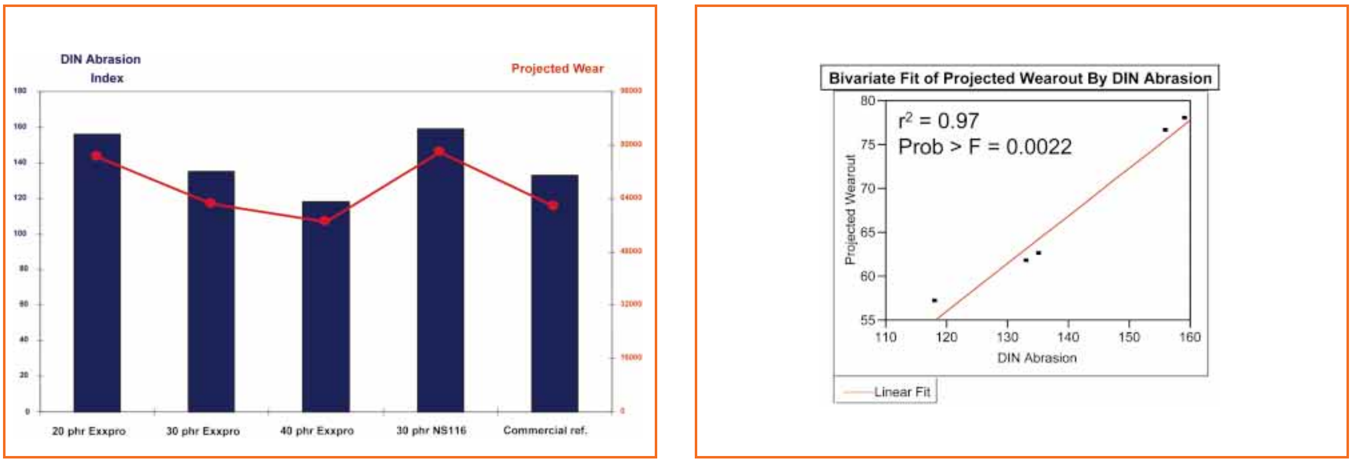

Fig. 9. SAS JMP analysis of projected wearout data from road wear testing at 95 km/h to laboratory DIN abrasion index values

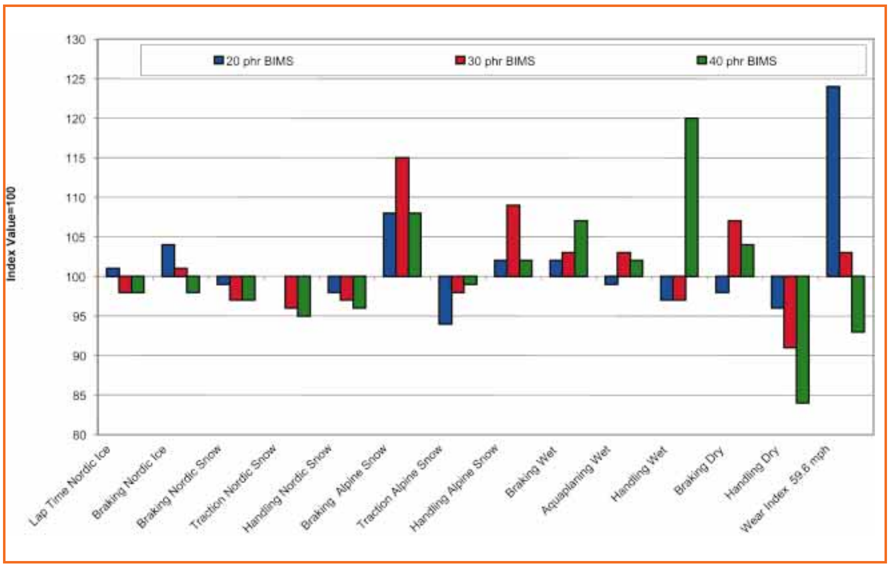

Fig. 10. Comparative summary of BIMS model winter tread tire performance normalized to that of the commercial reference compound

Summary

Vehicles having tires with BIMS model winter treads showed improvements indicating the potential of this elastomer for this application, see *Fig. 10*. At the three BIMS levels studied, shorter braking distances on an indoor ice surface, on an Alpine snow surface, and on wet and dry road surface conditions were obtained compared to the commercial reference or to the sSBR control compounds. The tires with 20-phr BIMS had comparable performance to tires with the 30-phr sSBR having a 20 % bound styrene level. Tires with 30-phr BIMS had improved traction on Alpine snow and wet asphalt surfaces. The improved braking and traction responses of tires with BIMS model treads are thought due to the second higher-temperature transition (above this Tg relaxation) due to local torsional motions along the BIMS polymer chain.

Dry handling was deficient, but was shown to correlate with the hardness of the BIMS model tread compounds whereby the softer treads afforded lower handling performance. BIMS model winter treads have projected wear-out values > 50,000 km showing their potential to last at least two seasons.

Vehicle performance for tires with BIMS treads was lower than expected on the ice and compacted frozen snow surfaces characteristic of Nordic winter conditions; hence BIMS use is not thought to be useful under these severe temperature conditions.

The ratios of BIMS, BR and NR, and the silica and silane levels need to be optimized in order to obtain specific tire performance advantages.

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