# Influence of Thermoplastic Polyurethane on the Properties of Asphalt Cement and the Elastic Behavior of Asphalt Mixtures

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#### **Abstract**

The useful life of pavements determines the design of asphalt mixtures. However, premature structural failures and low durability are one of the main problems in pavement engineering today. The action of loads produced by traffic, exposure to environmental factors and the inadequacy of conventional designs of asphalt pavements are some of the main problems associated with the durability of the pavement. The modification of asphalt binders is an alternative that has been used for some years to improve the properties of bituminous material. Among the modifying materials are elastomers, a class of polymers with elastic properties that can be heat-treated. In this study, the asphalt was modified by wet means by adding 5, 10 and 15% of Thermoplastic Polyurethane (TPU) by weight of the binder. Subsequently, standard samples were prepared with the characterized constituents and mixtures with the modified asphalt cement. Consequently, tests were carried out to obtain the optimal mixture that improves the properties of conventional pavement. The results show that the optimal TPU addition content is 5%. With this percentage, the modified binder presented a greater elastic recovery of up to 5.60% and a reduction of up to 42.6% in penetration compared to conventional asphalt. In addition, using the Marshall method, it was determined that the stability of the modified mixtures was much higher than that of standard mixtures for light, medium and heavy traffic; so, they will have a higher resistance to deformation and better fatigue behavior.

#### **Keywords**

modified asphalt, TPU, modified asphalt mix, wet process, Marshall Design

## 1 Introduction

The rapid deterioration of pavements, the high demands on vehicle loads, and the complexity of service conditions and requirements for road construction are the most significant challenges facing road engineering [1]. However, due to the heyday of the automotive industry and the lack of effective solutions to the recurrent traffic congestion in road systems that demarcate the current functioning of cities, the projection indicates that traffic conditions will worsen even more in the future, to the point that there will be perennially roads with insufficient capacity [2, 3].

The evolution of vehicular flow has an impact on the increase in traffic loads and, added to environmental conditions such as temperature variation and precipitation, facilitate the premature development of failures in the structure of asphalt pavements [4, 5]. Indeed, the factors associated with overloading are predominantly involved in the early phases of the pavement's life cycle [6].

For some years now, research has been carried out on the modification of asphalt with elastomeric polymers [7]. In general, the incorporation of these is beneficial for the binder because it allows to enhance properties such as elasticity, adhesion, cohesion, rheological and mechanical characteristics [8]. Additionally, the cost of incorporating these polymers as modifiers is offset by the increase in the useful life of the pavement [9].

One of the investigations [10] with this type of materials indicates that the addition of SBR (Styrene-Butadiene) polymer favored the resistance of asphalt mixtures under the action of humidity. Another polymer used in the modification of asphalt cement is SBS (Styrene-Butadiene-Styrene). In a study with the latter elastomeric polymer and the addition of Glisonite [11], it was determined that the penetration of the ligand was reduced with a higher SBS content and, therefore, showed an adverse effect on

the storage stability property. On the other hand, polypropylene as a modifier enhances the moisture resistance, adhesion and cohesion of the asphalt mixture, ensuring a better bond between the binder and the aggregates [12]. On the other hand, modification with nanomaterials does not greatly change the properties of conventional bituminous material, but it can increase its shelf life, improve rheological properties and storage stability, although it is an expensive material [13, 14].

TPU is a polymer with an internal structure made up of flexible and rigid segments that are joined together by covalent and hydrogen bonds [15]. It is part of the family of elastomers, which are characterized by being amorphous in their non-dilated state and by their elastic properties when they are at temperatures higher than that reached in their glass transition [16]. It originates from the interaction of three elements:

- 1. diisocyanates;
- 2. a linear chain diol that has a high molecular weight and can be considered a long-chain polyol, and
- 3. another diol that acts as a chain paver and has a low molecular weight [17].

It is a material with favorable conditions due to its reversible cross-linked structure that allows it to maintain its genuine natural characteristics and be thermally managed [18].

Previous studies with polyurethane in different presentations have proven the improvement in the behavior of bituminous materials and mixture, improving properties such as adhesion and stability at high temperatures [19]. The compatibility between polyurethane additives and PEN 60/80 asphalt under the molecular dynamics approach is optimal at a temperature of 135 °C, and the most significant cohesive energy density is achieved with 15% addition by weight of the binder [20]. Other recent research with TPU shows that the higher the amount of elastomer, the lower the penetration values of the modified binder, as well as an increase in properties such as ductility and softening point [21]. Likewise, another study indicates that failure resistance can be improved with the addition of 7% synthesized polyurethane [22].

### 2 Materials and methods

#### 2.1 Materials

The asphalt binder used in this research has a penetration rate of 60/70. Its properties are shown in Table 1.

Table 1 Properties of asphalt cement

Practice	Norm	Result
Penetration (100 g, 5 s, 25 °C), 0.1 mm	ASTM D5	61
Ductility (25 °C, 5 cm/min), cm	ASTM D113	100
Flash point, °C	ASTM D92	234
Solubility in trichloroethylene; %	ASTM D2042	99.8
Kinematic viscosity (135 °C)	ASTM D2170	205

The aggregates are characterized by being siliceous in nature, considering a grain size between 4.75 mm and 12.5 mm as a coarse aggregate (Table 2) and a range between values below 4.75 mm and greater than 0.075 mm as a fine aggregate (Table 3). The material used as an adhesion improver between aggregates-asphalt was Portland cement type I, with a grain size of less than 0.075 mm. Table 4 shows properties of TPU.

Table 2 Properties of coarse aggregate

Practice	Norm	Result
Abrasion the angels, %	ASTM C131	20.4
Flat, elongated particles, %	ASTM D4791	3.38
Fractured faces	ASTM D5821	85/50
Total soluble salts, %	LNV 8	0.45
Absorption %	ASTM C127	0.87

Table 3 Properties of fine aggregate

Practice	Norm	Result
Sand equivalent	ASTM D2419	61
Methylene blue	AASTHO TP57	1.6
Plasticity index (mesh No. 40)	ASTM D4318	NP
Plasticity index (mesh No. 200)	ASTM D 4318	NP
Total soluble salts	LNV 8	0.48
Absorption	ASTM C128	0.37

Table 4 Properties of TPU

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Property	Norm	Value		
Density, g/cm <sup>3</sup>	ISO 2781	1.2		
Shore hardness A, A	ISO 7619-1	84		
Tensile strength, MPa	ISO 22654	58.2		
Elongation at break, %	ISO 22655	570		
Module 100%, MPa	ISO 22656	5.2		
Module 300%, MPa	ISO 22657	14.8		
Tear strength (angle), N/mm	ISO 34-1	83		
Permanent deformation at 23 °C, %	ISO 815-1	22		
Abrasion resistance, mm	ISO 4649	35		
Vicat softening point, °C	ISO 306	90		

#### 2.2 Methods

# 2.2.1 Sample preparation

The method of interest in this study is wet ligand modification using a laboratory colloidal mill. The percentages of addition considered were 5, 10 and 15% TPU by weight of the binder.

The natural state of conventional asphalt is solid at room temperature. For the modification, asphalt cement was placed in the kiln and heated to a temperature of 175 °C for approximately 10 minutes, until the appropriate degree of fluidity was obtained. In each experience of asphalt modification with TPU in different proportions by weight of the binder, the asphalt cement sample was 5500 g. The weight of the polymer was determined (Fig. 1). Table 5 describes the range of application.

## 2.2.2 Asphalt cement modification

The pure asphalt binder was mixed with the elastomeric polymer in a laboratory colloidal mill. The sample, with the appropriate degree of fluidity, remained inside the equipment at a temperature of 175 °C. Subsequently, the TPU was slowly placed in fractional contents for each defined percentage, until it could be verified that the asphalt-polymer mixture was homogeneous.

The materials were mixed at a speed of 3500 rpm over the course of 40 minutes. The path of the bitumen together with the polymer took place on the surface enclosed by a grooved conical rotor at its periphery and a grooved stator, also conical, which remains fixed.



Fig. 1 Elastomeric polymer TPU

Table 5 Weight of polymer at each dosage by weight of asphalt

Contents of TPU (%)	Weight (g)
5	275
10	550
15	825

The high-shear equipment has an adjustment device that allowed the fineness of the thermoplastic elastomer to be handled without altering its operation, so that the modified asphalt does not present traces of the polymer (Fig. 2).

## 2.2.3 Asphalt mix design

The asphalt mix with conventional asphalt cement and TPU modified asphalt was designed using the Marshall method in the laboratory.

The aggregates used for the asphalt mixtures complied with the minimum requirements established in the national regulations of the General Specifications of the MTC [23]. For this research, the corresponding conditions for aggregates were defined for areas with altitude less than 3000 m.a.s.l.

Various mixtures of coarse and fine aggregates were tested with different percentages was performed to establish the working formula and classify the samples according to the aggregate gradation parameters for MAC 2 (Fig. 3) according to [23]. Table 6 shows the results obtained.



Fig. 2 Disposal of TPU-modified asphalt cement

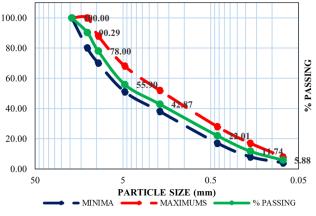


Fig. 3 Gradation MAC 2

Table 6 Determining MAC Parameters

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Sieve	Aperture (mm)	% Intern	Specs	MAC 2	
1"	25.4	100			
3/4"	19.1	100	100	100	
1/2"	12.5	90.29	80	100	
3/8"	9.5	78	70	88	
1/4"	6.35	66.99			
N° 4	4.75	55.9	51	68	
N° 8	2.36	46.86			
N° 10	1.9	42.87	38	52	
N° 16	1.19	35.32			
N° 30	0.6	26.78			
N° 40	0.42	22.01	17	28	
N° 50	0.3	16.85			
N° 80	0.177	11.74	8	17	
N° 100	0.15	10.05			
N° 200	0.075	5.88	4	8	
Fondo		0			

For the design of asphalt mixtures using the Marshall method, the criteria indicated by the Asphalt Institute were considered [24]. Four initial asphalt contents (4.5, 5, 5.5 and 6%) were established and through the results of the test, considering the maximum stability, the highest unit weight and a percentage of voids as shown in the parameters for each type of traffic, the optimal content was determined. Table 7 shows the working formula established for the design. Table 8 shows the criteria of the asphalt institute.

Table 7 Working formula for the design of asphalt mixtures

Asphalt content/A. modified	4.5	5	5.5	6
Percentage of coarse aggregate	47.5	47.5	47.5	47.5
Fine aggregate percentage	50	50	50	50
Filler percentage	2.5	2.5	2.5	2.5
Total	100	100	100	100

Table 8 Criteria for Marshall Design

Marshall method mixing criteria	Light	transit		lium nsit	Heavy	traffic
	Min	Max	Min	Max	Min	Max
Compaction (number of strokes on each side)	3	5	5	0	7	75
Stability, N	3336		5338		8006	
Flow, 0.25 mm (0.1")	8	18	8	16	8	14
Percentage of voids	3	5	3	5	3	5
Percentage of VMA (TM <sup>3</sup> / <sub>4</sub> ")	12 14		12 14		12 14	
Percentage VFA	70	80	65	78	65	75

#### 3 Results and discussions

#### 3.1 Results

This study was carried out in two well-defined phases:

- 1. the addition of TPU as a modifier of asphalt cement and
- 2. the design of asphalt mixtures with the modified binder.

The experimental design is presented in Fig. 4. The properties of modified asphalt cement are described in the following sections.

#### 3.1.1 Penetration

The penetration results of TPU-modified asphalt show an inversely proportional relationship with respect to the polymer content, with a decreasing trend. The difference between what was obtained with conventional asphalt and the highest percentage of addition considered in this research was 31 mm, as shown in Fig. 5.

# 3.1.2 Ductility

It was determined that the ductility of the modified asphalt cement presents values close to that of the pure binder, showing a slight increase of only 2 cm with respect to the maximum addition content of TPU, as shown in Fig. 6.

# 3.1.3 Torsional elastic recovery

The torsional elastic recovery of asphalt with a 60/70 degree of penetration was nil. The results show that the addition of TPU increases the recovery rate by up to 6.70% as shown in Fig. 7.

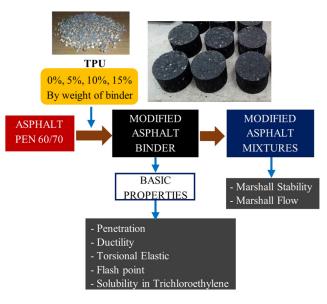


Fig. 4 Experimental design flow chart

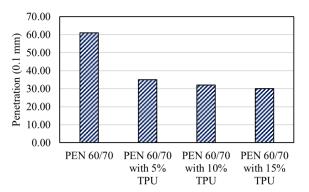


Fig. 5 Penetration of conventional and modified asphalt

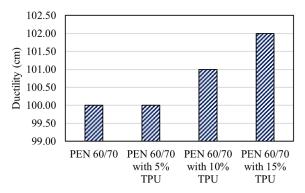
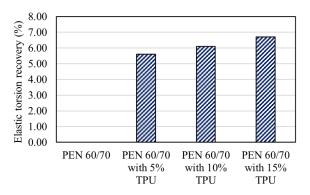


Fig. 6 Ductility of conventional and modified asphalt



 $\textbf{Fig. 7} \ \textbf{Torsional elastic recovery of conventional and modified as phalt}$ 

# 3.1.4 Flash point

This property shows values with an increasing trend as the content of the elastomeric polymer increases (Fig. 8).

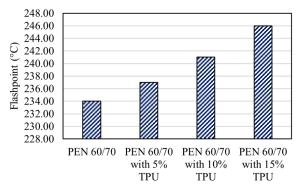


Fig. 8 Flash point of conventional and modified asphalt cement

## 3.1.5 Solubility in trichloroethylene

The purity of conventional asphalt cement remained constant even with the addition of 5 and 10% TPU (as evidenced in the Fig. 9) with a value of 99.80%. Meanwhile, with the maximum dosage of the polymer by weight of the binder, it decreased by 0.10% in accordance with the above.

#### 3.1.6 Marshall stability

It was determined that the ability to resist deformation increased significantly as the content of the thermoplastic elastomer increased, compared to conventional asphalt mix. In the light transit type, stability went from 1460 kg to 1725, 1760 and 2000 kg, for percentages of 5, 10 and 15% TPU.

For the medium transit the results followed the same tenancy, the Marshall stability of the conventional mixture was 1450 kg, while for the asphalt mixtures with modified binder it was 2030, 2080 and 2390 kg for the percentages considered in ascending order.

With respect to heavy traffic, the maximum stability for asphalt mix with a penetration degree of 60/70 was 1369 kg. In addition, for mixtures with modified binder with the addition of 5, 10 and 15% TPU, the Marshall stability achieved was 2350, 2450 and 2570 kg.

## 3.1.7 Marshall flow

In the case of conventional asphalt mix in light traffic, a flow of 4.5 mm was reached. Mixtures with the 5 and 10% modified binder reduced this value to 4.45 and 4.29 mm, respectively. While with 10% TPU 4.71 mm was obtained.

Regarding the medium traffic, the flow in the conventional mixture was 3.9 mm; On the other hand, for additions of 5, 10 and 15% TPU to the binder, the fluence achieved was 3.51, 4.14 and 3.3 mm.

For heavy traffic mixtures, the initial flow obtained was 3.47 mm. On the other hand, for mixtures with modified binder, the creep values were: 4.06, 3.68 and 3.68 mm,

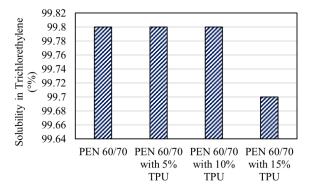


Fig. 9 Trichloroethylene solubility of conventional and modified asphalt

respectively, for the percentages of additions considered in this research.

Tables 9–12 show the results of the Marshall test for conventional asphalt mix and TPU-modified binder.

Table 9 Properties of asphalt mix with conventional asphalt

Transit type	Light	Medium	Heavy
Optimal content C.A. (%)	5.8	5.4	5.5
Unit weight (g/cm2)	2.246	2.23	2.242
Empty (%)	4	3.2	3.05
Mineral aggregate voids (%)	21.6	21.7	21.31
Voids filled with C.A. (%)	79	86	85.31
Flow (mm)	4.5	3.9	3.47
Stability (kg)	1460	1450	1369
Dust-to-asphalt ratio	0.98	0.91	0.94

Table 10 Properties of asphalt mix with asphalt cement modified with 5% TPU

Transit type	Light	Medium	Heavy
Optimal content C.A. (%)	5.8	5.7	5.2
Unit weight (g/cm2)	2.25	2.27	2.27
Empty (%)	4	3.4	4
Mineral aggregate voids (%)	21.2	20.4	20.2
Voids filled with C.A. (%)	77	83	80
Flow (mm)	4.45	3.51	4.06
Stability (kg)	1725	2030	2350
Dust-to-asphalt ratio	0.98	0.97	0.88

Table 11 Properties of asphalt mix with 10% TPU modified asphalt cement

Transit type	Light	Medium	Heavy
Optimal content C.A. (%)	5.3	5.2	5.4
Unit weight (g/cm2)	2.242	2.265	2.74
Empty (%)	3	4	3.3
Mineral aggregate voids (%)	21.2	20.4	20.2
Voids filled with C.A. (%)	86	80	84
Flow (mm)	4.29	4.14	3.68
Stability (kg)	1760	2080	2450
Dust-to-asphalt ratio	0.9	0.88	0.92

Table 12 Properties of asphalt mix with 15% TPU modified asphalt cement

Transit type	Light	Medium	Heavy
Optimal content C.A. (%)	5.5	5.2	5.4
Unit weight (g/cm2)	2.21	2.241	2.23
Empty (%)	3.2	2.15	3
Mineral aggregate voids (%)	22.45	21.3	21.9
Voids filled with C.A. (%)	87	90	90
Flow (mm)	4.72	3.3	3.68
Stability (kg)	2000	2390	2570
Dust-to-asphalt ratio	0.94	0.88	0.92

#### 3.2 Discussions

According to the tests carried out in the first stage of the research, it was determined that with the addition of TPU the ductility values at a temperature of 25 °C do not show significant changes. In general, it shows a slow increase exceeding the minimum value of 100 cm of the pure binder, reaching a maximum of 102 cm with 15% addition. In contrast to Zhang et al. [21], ductility values also increase as TPU content increases; however, up to 11% of addition, ductility was less than 100 cm, a limit that was later exceeded with percentages of 13 and 15% TPU. However, Jin et al. [25] evaluated the ductility at low temperature (5 °C) of an asphalt with a penetration degree of 80/100 modified with polyurethane (PU) and asphalt rock (RA) composite, and the results were positive, with values close to 12.5 and 21 cm for 5 and 10% PU (0% RA), which compared to conventional (8.6 cm) represent an increase of 45 and 144%. respectively. In another research [26], it was determined that with SBS polymer plus organic bentonite, modified asphalt had a 210% increase in ductility over conventional asphalt.

The results of the torsional elastic recovery test of the modified asphalt binder have a positive trend directly proportional to the increase in elastomer content, reaching values of 5.6 and 6.7% for TPU additions of 5 and 10%. In contrast to the study of [27], with the modification of the binder with PU by means of the in-situ synthesis methodology, obtaining elastic recovery results at high temperature (0.1 KPa) greater than 20% with 3% addition, and exceeding 70% recovery with 6% PU.

The penetration test (0.1 mm) of the modified asphalt showed that this property is reduced with a greater amount of TPU, being 30 mm with the maximum percentage of addition. The results agree with those obtained in the study of [21], in which penetration values below 60 mm are detailed starting from 1% addition, obtaining with 15% TPU a minimum value slightly higher than 52.5 mm. In another study [28], TPU addition percentages of 3, 5 and 7% were applied to a pure binder with a penetration rate of 80/100, obtaining values of 70, 55 and 45 mm for the indicated contents. This shows that the higher the polymer content, the lower the penetration of the binder.

Regarding the flash point of the binder, it is detailed in [23] that the minimum temperature, both for asphalt with a penetration degree of 60/70 and modified with polymers of different types, in which the necessary conditions are present in which the bituminous material is susceptible to ignition by the action of an ignition source is

232 °C. With the addition of TPU in percentages of 5, 10 and 15%, the flash point results were 237, 241 and 246 °C, respectively. In all cases, the minimum parameter established in the standard was complied with.

In one study [29] they were commissioned to analyze the properties of polymer-modified asphalt cement (PMB) with 1% addition. The results show a reduction in penetration from 64.4 to 57 mm; the decrease in ductility from 128.5 to 112 cm and the improvement of the minimum inflation temperature from 255 to 271 °C.

Through the solubility test in trichloroethylene, it was shown that the purity of the binder remains constant to the conventional one even with the addition of 5 and 10% TPU, exceeding the minimum requirement of 99% set out in [23] for asphalts with different degrees of penetration and for polymer modified asphalts. In the case of 15% TPU, solubility was significantly reduced, so it remained compliant.

In the second stage, it was determined that in the C mixture (light transit) the Marshall stability increases significantly with each percentage of addition, surpassing that obtained in the mixture with the pure binder 60/70 which was 1460 kg for an optimal asphalt content of 5.8%. Based on the parameters described in [24], the engineered mixtures exceed the minimum stability of 340 kg (3336 N) and the creep within the range of 2.0 and 4.6 mm. Additionally, the flows had values close to the asphalt mix without addition with 4.50 mm. However, with 15% TPU the Marshall flux obtained is not within the range.

For the medium transit type, the stability values significantly exceeded the minimum value specified in [24], which is 544 kg (5338 N). In addition, the flow values had to be in a range of 2 to 4.1 mm, note that only the mixture with 5 and 15% TPU meet this parameter with 3.51 and 3.3 mm, correspondingly.

The mixtures for the heavy traffic type showed a strain resistance capacity much higher than the criteria of [24], where the minimum stability value is 816 kg (8006 N) and the Marshall flow in the range of 2 to 3.6 mm. TPU-modified mixtures from heavy traffic did not meet this last parameter, as fluences greater than 3.6 mm were obtained.

In a study by Hong et al. [30] they employed TPU and amorphous polyalpha olefins (APAO). A maximum stability of more than 12 kN (1224 kg) and a Marshall flow slightly greater than 3 mm were obtained with a ratio of 2% TPU + 6% APAO by weight of the binder. In addition, when only the binder was modified with 2% TPU, the stability achieved was 10 kN and the flow rate was

approximately 4 mm. Another research by Zhang et al. [31], when measuring strain resistance, obtained a Marshall stability of 11.2 kN with 2% SBS polymer, while with 4% of the polymer the Marshall stability was 12.17 kN, lower values compared to TPU-modified mixtures.

#### 4 Conclusions

The present research studied the effects of the modification of asphalt cement with the addition of TPU on its basic properties and the elastic behavior of wet-modified asphalt mixtures as a continuation of the first phase.

Based on the results, it was ruled out that the optimal mix is with 10% TPU because the Marshall flow was only within the parameters in the light traffic type; while with 15%, mixtures with excess rigidity were obtained as they showed very high stability. Thus, it was determined that the best addition content is 5% because it allows an increase in stability and power, like the other percentages of addition, the properties of asphalt cement. The analysis of the results and conclusions of this study is detailed below.

The penetration property of modified asphalts shows substantial changes. The results of the test indicate that it was reduced from 61 mm to 35, 32 and 30 mm for 5, 10 and 15% TPU, which shows that the elastomeric polymer makes the asphalt harder and thus improves stability at high temperatures.

The ductility of the modified binders does not change significantly. The values obtained were 100 cm, 101 cm and 102 cm for elastomer contents of 5, 10 and 15%, which compared to the ductility of asphalt with a penetration degree of 60/70 which was 100 cm, does not show an improvement in the ductile response.

The torsional elastic recovery of the experimental group took values of 5.6, 6.1 and 6.7% for proportions of 5, 10 and 15% addition. On the other hand, that of the virgin binder is considered null. This difference shows that the modified binder mixture will have better fatigue behavior than conventional mixtures, but not as largely expected.

Regarding the flash point, good results were obtained as the content of the modifier was increased. This leads to the modified binder becoming flammable at minimum temperatures higher than that of the conventional binder, which was 234 °C.

Through the Trichloroethylene solubility test, it was demonstrated that the TPU-modified binder maintains its purity with a composition of more than 99% active binding material and with a minimum amount of contaminants in all experimental groups.

The Marshall test showed that stability is higher in all experimental groups than in the control group, indicating a greater ability to resist deformation in the face of the action of traffic loads. Likewise, the flow of the modified mixtures was close to that of the conventional mixture; however, mixtures designed for heavy traffic modified with TPU showed Marshall flows that did not comply with regulations.

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