UNIVERSIDAD CATÓLICA SANTO TORIBIO DE MOGROVEJO

FACULTAD DE INGENIERÍA

ESCUELA DE INGENIERÍA CIVIL AMBIENTAL

Influence of thermoplastic polyurethane on the properties of asphalt cement and the elastic behavior of asphalt mixtures

TRABAJO DE INVESTIGACIÓN PARA OPTAR EL GRADO ACADÉMICO DE BACHILLER EN INGENIERÍA CIVIL AMBIENTAL

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Chiclayo, 2023

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Resumen

La vida útil de los pavimentos es un factor importante para el diseño de las mezclas asfálticas. Sin embargo, las fallas prematuras en su estructura y la baja durabilidad constituyen uno de los principales problemas de la ingeniería de pavimentos actualmente. La causa esencial que se atribuye es la deformación constante sobre sus capas generadas por el incremento de la acción de las cargas tráfico. La modificación de ligantes asfálticos es una alternativa que se viene empleando desde hace algunos años y tiene como objetivo la mejora de las propiedades del material bituminoso y la durabilidad de los pavimentos asfálticos. Dentro de los materiales modificadores se encuentran los elastómeros, que son una clase de polímeros con propiedades elásticas que se pueden tratar térmicamente. En este estudio se evaluaron las propiedades básicas del asfalto modificado mediante la adición de 5, 10 y 15% de Poliuretano Termoplástico (TPU) en peso del ligante y el comportamiento elástico de las mezclas asfálticas para tránsito liviano, mediano y pesado por el método Marshall empleando el cemento asfaltico modificado. Se demostró que las propiedades de recuperación elástica torsional y punto de inflamación del asfalto modificado mostraron una tendencia positiva creciente, otras como la ductilidad y la solubilidad no sufrieron cambios relevantes, a diferencia de la penetración que se redujo mientras se aumentaba el contenido del polímero. Adicionalmente, se determinó que las mezclas asfálticas tendrán una mayor resistencia a la deformación y un mejor comportamiento a la fatiga, pero no de forma significativa.

Palabras clave: Asfalto Modificado, Poliuretano Termoplástico, Mezcla Asfáltica Modificada, Proceso Húmedo, Diseño Marshall.

Abstract

The service life of pavements is an important factor for the design of asphalt mixes. However, premature structural failures and low durability are one of the main problems in pavement engineering today. The essential cause attributed to it is the constant deformation on its layers generated by the increase in the action of traffic loads. The modification of asphalt binders is an alternative that has been used for some years and aims to improve the properties of the bituminous material and the durability of asphalt pavements. Among the modifying materials are elastomers, which are a class of polymers with elastic properties that can be heat-treated. In this study, the basic properties of modified asphalt were evaluated by adding 5, 10 and 15% Thermoplastic Polyurethane (TPU) by weight of the binder and the elastic behavior of asphalt mixtures for light, medium and heavy traffic by the Marshall method using modified asphalt cement. It was shown that the torsional elastic recovery and flash point properties of the modified asphalt showed an increasing positive trend, others such as ductility and solubility did not undergo relevant changes, unlike penetration which was reduced while the polymer content was increased. Additionally, it was determined that asphalt mixtures will have a higher resistance to deformation and a better fatigue behavior, but not significantly.

Keywords: Modified Asphalt, Thermoplastic Polyurethane, Modified Asphalt Mix, Wet Process, Marshall Design.

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 current traffic congestion, the projection indicates that traffic conditions will worsen further in the future [2].

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 [3, 4]. Indeed, the factors associated with overloading are predominantly involved in the early phases of the pavement's life cycle [5].

For some years now, research has been carried out on the modification of asphalt with elastomeric polymers [6]. 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 [7]. Additionally, the cost of incorporating these polymers as modifiers is offset by the increase in the useful life of the pavement [8].

One of the investigations [9] 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 [10], 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 [11]. 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 [12, 13].

Thermoplastic Polyurethane (TPU) is a polymer with an internal structure made up of flexible and rigid segments that are joined together by covalent and hydrogen bonds [14]. 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 [15]. 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 [16]. 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 [17].

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 [18]. 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 [19]. 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 [20]. Likewise, another study indicates that failure resistance can be improved with the addition of 7% synthesized polyurethane [21].

Materials and methods

Materials

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

PRACTICE	VALUE	EG 2013
Penetration (100g, 5s, 25°C), 0.1mm	61	60 - 70
Ductility (25°C, 5cm/min), cm	100	100 mín.
Flash point, °C	234	232 mín.
Solubility in Trichloroethylene; %	99.8	99 mín
Kinematic viscosity (135°C)	205	200 mín.

Table 1. Properties of asphalt cement

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

PRACTICE	VALUE	EG 2013
Absorption, %	0.87%	1% máx.
Abrasion The Angels, %	20.40%	40% máx.
Total Soluble Salts, %	0.45%	0.5% máx.
Flat and elongated particles, %	7.33%	10% máx.
Fractured faces	89/86	85/50

Table 2. Properties of Coarse Aggregate

FINE AGGREGATE	VALUE	EG 2013
Methylene Blue	1.6	8 máx.
Absorption, %	0.37%	0.5% máx.
Total Soluble Salts, %	0.48%	0.5% máx.
Plasticity Index (mesh N°40)	NP	NP
Plasticity Index (mesh N° 200)	NP	4 máx.

Table 3. Properties of Fine Aggregate

Table 4 shows properties of thermoplastic polyurethane:

PROPERTY	NORM	VALUE
Density, gr/cm3	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

Table 4. Properties of Thermoplastic Polyurethane

Methods

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 gr. The weight of the polymer was determined. Table 5 describes the range of application.

CONTENTS OF TPU (%)	WEIGHT (gr.)
5	275
10	550
15	825

Table 5. Weight of Polymer at Each Dosage by Weight of Asphalt

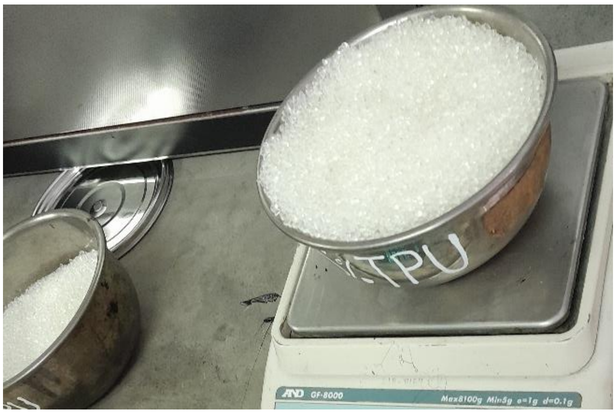

Fig. 1. Elastomeric polymer Thermoplastic polyurethane (TPU)

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.

Fig. 2. Disposal of TPU-modified asphalt cement

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.

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.

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 [22]. 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 according to [22]. Table 6 shows the results obtained:

SIEVE	APERTURE (MM)	% PASSING	SPECS MAC 2	
1”	25.4	100		
¾”	19.1	100	100	100
½”	12.5	90.29	80	100
⅜”	9.5	78	70	88
¼”	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
Bottom		0		

Table 6. Determining MAC Parameters

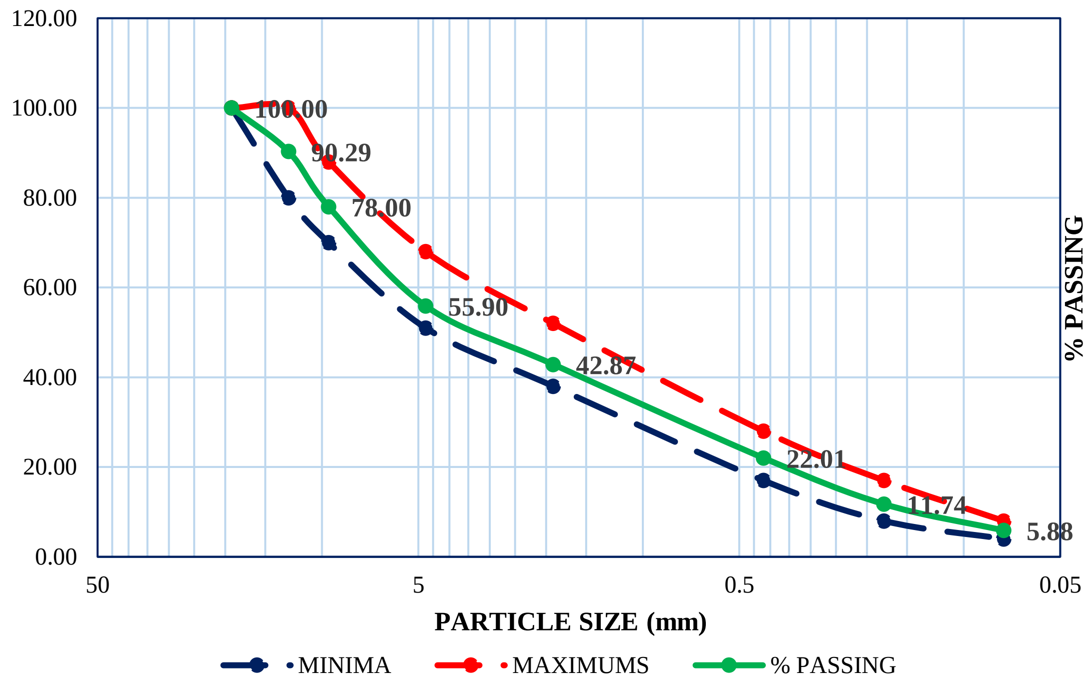

Fig. 3. Gradation MAC 2

For the design of asphalt mixtures using the Marshall method, the criteria indicated by the Asphalt Institute were considered [23]. 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:

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 7. Working Formula for the Design of Asphalt Mixtures

MARSHALL METHOD MIXING CRITERIA	LIGHT TRANSIT		MEDIUM TRANSIT		HEAVY TRAFFIC	
	MIN	MAX	MIN	MAX	MIN	MAX
Compaction (number of strokes on each side)	35		50		75	
Stability, kN	4.53		5.44		8.15	
Flow, 0.25mm (0.1”)	8	20	8	16	8	14
Percentage of voids	3	5	3	5	3	5
Percentage of VMA (TM ¾”)	14		14		14	
Dust - Asphalt ratio	0.6	1.3	0.6	1.3	0.6	1.3

Table 8. Criteria for Marshall Design

Results and discussion

Results

This research was divided into two stages: (1) the addition of TPU as a modifier to the asphalt binder and (2) the hot design of the asphalt mixture with the modified binder. The properties of the modified asphalt cement were:

a) 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. 4.

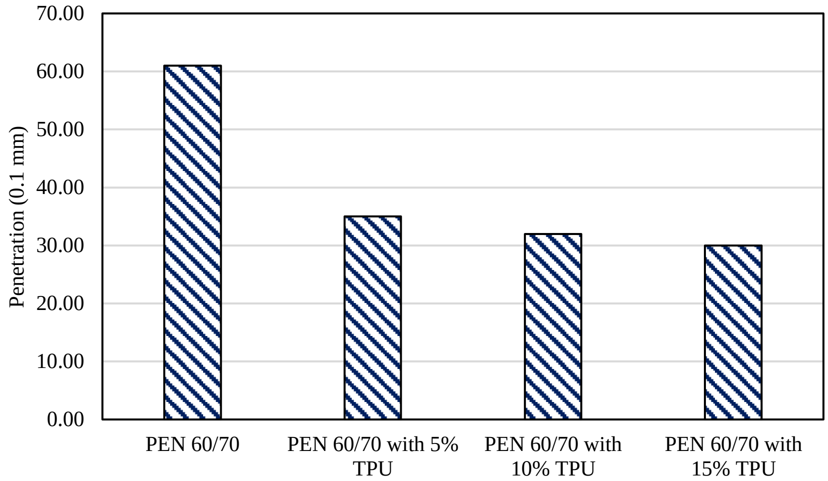

Fig. 4. Penetration of conventional and modified asphalt cement

b) 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. 5.

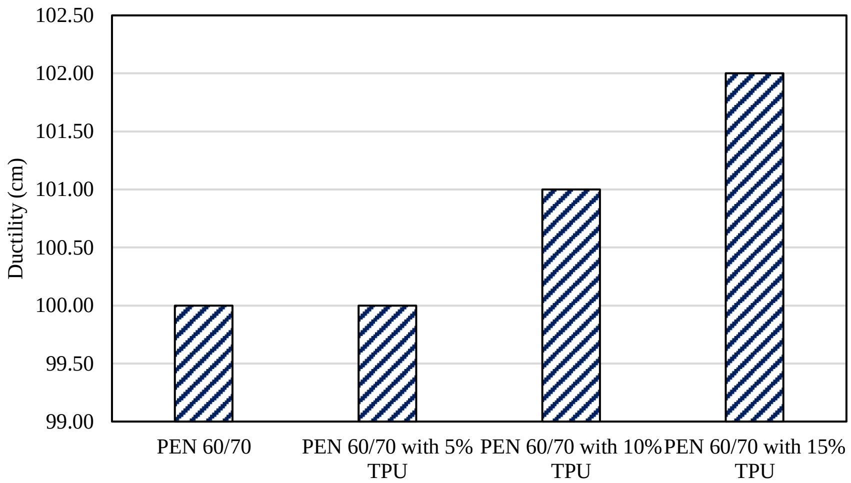

Fig. 5. Ductility of conventional and modified asphalt cement

c) 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. 6.

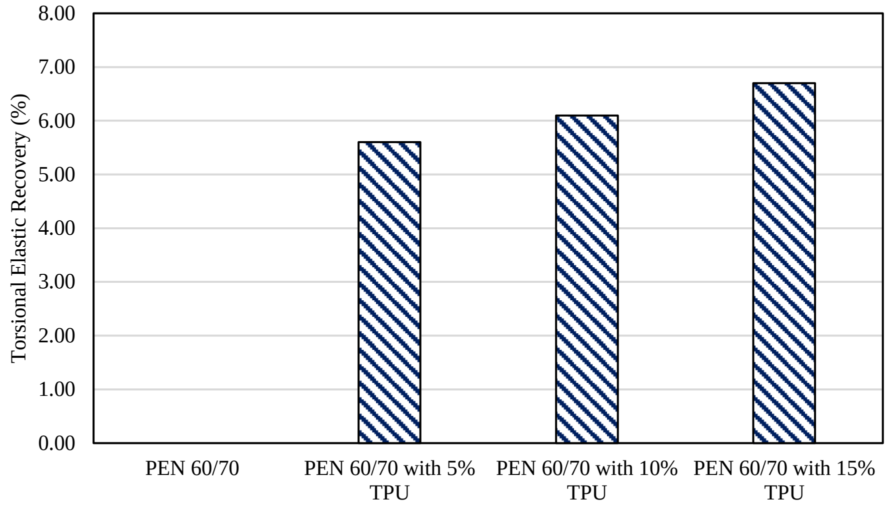

Fig. 6. Torsional elastic recovery of conventional and modified asphalt cement

d) Flash Point

This property shows values with an increasing trend as the content of the elastomeric polymer increases, as shown in Fig. 7.

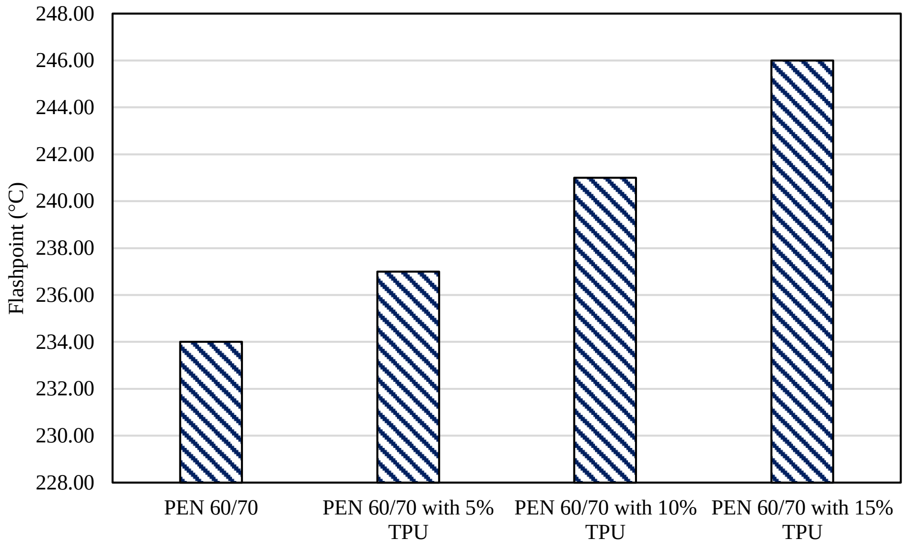

Fig. 7. Flash point of conventional and modified asphalt cement

e) 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. 8) 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.

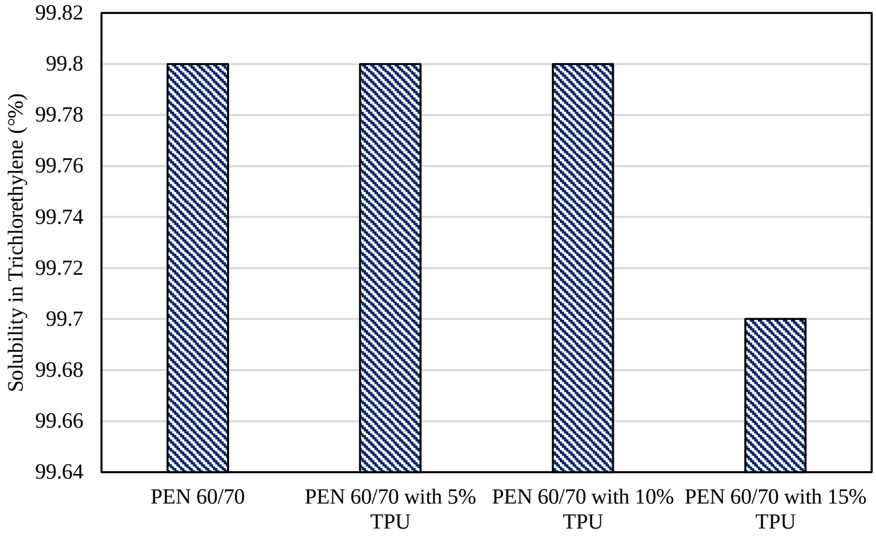

Fig. 8. Trichloroethylene solubility of conventional and modified asphalt cement

f) Marshall stability

It was determined that the resistance to pitting increased significantly as the content of the thermoplastic elastomer increased, compared to the conventional asphalt mixture. 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 resistance to permanent deformation for 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 the asphalt mix with a penetration degree of 60/70 was 1369 kg. Additionally, for mixtures with modified binder with 5, 10 and 15% TPU addition, the denting resistance they achieved was 2350, 2450 and 2570 kg.

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

Tables 9, 10, 11 and 12 show the results of the Marshall test for conventional asphalt mix and TPU-modified binder.

TRANSIT TYPE	LIGHT	MEDIUM	HEAVY
Optimal content C.A. (%)	5.8	5.4	5.5
Unit Weight (gr/cm2)	2.246	2.23	2.242
Empty (%)	4	3.2	3.05
Mineral Aggregate Voids (%)	21.6	21.7	21.31
Flow (mm)	4.5	3.9	3.47
Stability (kg)	1460	1450	1369
Dust-to-Asphalt Ratio	0.98	0.91	0.94

Table 9. Properties of asphalt mix with conventional asphalt

TRANSIT TYPE	LIGHT	MEDIUM	HEAVY
Optimal content C.A. (%)	5.8	5.7	5.2
Unit Weight (gr/cm2)	2.25	2.27	2.27
Empty (%)	4	3.4	4
Mineral Aggregate Voids (%)	21.2	20.4	20.2
Flow (mm)	4.45	3.51	4.06
Stability (kg)	1725	2030	2350
Dust-to-Asphalt Ratio	0.98	0.97	0.88

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

TRANSIT TYPE	LIGHT	MEDIUM	HEAVY
Optimal content C.A. (%)	5.3	5.2	5.4
Unit Weight (gr/cm2)	2.242	2.265	2.74
Empty (%)	3	4	3.3
Mineral Aggregate Voids (%)	21.2	20.4	20.2
Flow (mm)	4.29	4.14	3.68
Stability (kg)	1760	2080	2450
Dust-to-Asphalt Ratio	0.9	0.88	0.92

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

TRANSIT TYPE	LIGHT	MEDIUM	HEAVY
Optimal content C.A. (%)	5.5	5.2	5.4
Unit Weight (gr/cm2)	2.21	2.241	2.23
Empty (%)	3.2	2.15	3
Mineral Aggregate Voids (%)	22.45	21.3	21.9
Flow (mm)	4.72	3.3	3.68
Stability (kg)	2000	2390	2570
Dust-to-Asphalt Ratio	0.94	0.88	0.92

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

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. [20], 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, Xin Jin et al. [24] 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 [25], 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 [26], with the modification of the binder with polyurethane 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% polyurethane.

The penetration test (0.1mm) 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 [20], 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 [27], 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 [22] 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 [28] 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 [22] 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 [23], 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 4.50mm. 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 [23], 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.

Mixtures for the heavy traffic type showed a pitting resistance far superior to the criteria of [23], in which the minimum stability value was 816 kg (8006 N) and the Marshall flow in a range of 2 to 3.6 mm. For no TPU addition, this last parameter was met, since fluences above 3.6 mm were obtained.

In a study conducted by Hong et al. [29] They employed Thermoplastic Polyurethane (TPU) and Poly Alpha Amorphous Olefins (APAO). A maximum stability of more than 12 kN (1224 kg) and a flow slightly above 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 [30], when measuring the resistance by permanent deformation, 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, which are lower values compared to TPU-modified mixtures.

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.

The properties of penetration, ductility, torsional elastic recovery, flash point, solubility in trichloroethylene and Marshall parameters of the TPU-modified mixtures were evaluated and contrasted with the standard mixture. Based on this, it was determined that the best addition content of TPU is 5%. 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 61mm to 35, 32 and 30mm 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 better resistance to deformation. Likewise, the flow of the modified mixtures was close to the conventional mixture; however, in some percentages it did not comply with the regulations.

Acknowledgement

The authors express their deepest gratitude to the Universidad Católica Santo Toribio de Mogrovejo for facilitating access to the Pavement laboratories.

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