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Bitumen modified with recycled polyurethane foam for employment in hot mix asphalt

Betún modificado con espuma de poliuretano reciclada para empleo en mezclas bituminosas en caliente

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ABSTRACT

A wide variety of modifiers have been applied to bitumen in order to enhance their properties and performance. Among them, polymers have been mainly used. The aim of this paper is to assess the use of polyurethane foam waste as a bitumen modifier for hot mix asphalts. The polyurethane foam is a by-product of the manufacturing of polyurethane for thermal insulation. From a bitumen with a penetration grade of 50/70, various samples with percentages of waste material in weight ranging from 1% to 5% were produced and tested. Samples with 5% of waste material or more became rough and were refused due to their poor workability. A bituminous mixture with modified bitumen with a 4% of polyurethane was manufactured and compared with a sample with the same aggregates and original bitumen. Results in Marshall test showed that a mix with polymer modified bitumen yielded improvements in stability and a lower deformability. This result suggests that the employment of polyurethane foam waste is a promising bitumen modifier, contributing also to recycle waste materials.

Keywords: polyurethane foam, waste product, bitumen modifier, hot mix asphalt, polymer modified bitumen

RESUMEN

Una gran variedad de modificadores se han aplicado al betún para mejorar sus propiedades y rendimiento. Entre ellos, los polímeros han sido principalmente utilizados. El objetivo de este artículo es evaluar el uso del residuo de espuma de poliuretano como modificador de betún en mezclas bituminosas en caliente. La espuma de poliuretano es un residuo que se obtiene durante la fabricación del poliuretano para aislamiento térmico. A partir de un betún de penetración 50/70, se fabricaron y analizaron varias muestras con porcentajes de residuo en peso entre el 1 y el 5%. Las muestras con porcentajes del 5% de residuo o superiores produjeron un material áspero y se descartaron debido a su poca trabajabilidad. Se fabricó una mezcla bituminosa con un 4% de porcentaje de betún modificado con residuo de poliuretano y fue comparado con una muestra con los mismos áridos y el betún original. Los resultados en el test de Marshall mostraron que la mezcla con betún modificado provocaba un aumento en la estabilidad y una menor deformación. Estos resultados sugieren que el empleo de residuo de espuma de poliuretano es un prometedor modificador de betún, contribuyendo además al reciclado de materiales de desecho.

Palabras clave: espuma de poliuretano, residuo, modificador de betún, mezcla bituminosa en caliente, betún modificado con polímeros.

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Introduction

Bituminous asphalt mixtures are composed of bitumen, coarse and fine aggregates and mineral filler. Bitumen is a dark brown to black material principally obtained from crude oil distillation. It is a complex material, basically formed by hydrocarbons along with some other molecules containing small percentages of heteroatoms (sulphur, nitrogen and oxygen) (Cuadri *et al.*, 2014). From the point of view of the solubility in *n*-hetpane, the types of molecular species existing in the bitumen are classified into two main fractions: asphaltenes and maltenes. The asphaltenes are composed of highly condensed planar and heteroatom polar groups, polar aromatic ring systems and large amounts of heteroatom polar functional groups (Dong *et al.*, 2005). Maltenes are divided into three groups according to the solubility and polarity: saturates, aromatics and resins (Redelius, 2000). Hence, physico-chemical and rheological properties of

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the bitumen are dependent on both temperature and the relative proportion of mentioned fractions: asphaltenes, saturates, aromatics and resins (Fernández-Gómez *et al*., 2013). Traditionally, a colloidal model is used to describe bitumen behaviour, where asphaltenes are dispersed into an oily matrix of maltenes and peptized by resins (Lesueur, 2009). Due to its properties (ductility, waterproofing, adhesiveness, mechanical resistance, etc.), bitumen is generally employed in road pavement as a binder of mineral aggregates (Airey, 2003).

The traditional production procedure of asphalt mixture is carried out at high temperatures, above 140 °C, to assure that the low viscosity of the bitumen coats the aggregates. Mixes carried out following this technique are called Hot Mix Asphalts (HMA) (Rondón-Quintana *et al*., 2015). Alternative technologies have emerged during the last decades, such as the cold-mix asphalts (temperatures below 60 °C). Other techniques are Half-Warm-Mix Asphalt (temperatures from 60 °C to 100 °C) and Warm-Mix Asphalt (temperatures from 100 °C to 140 °C) (Rondón-Quintana *et al.,* 2016)

Bitumen is the only deformable component of the asphalt mixes, forming the continuous matrix of them (Adedeji *et al*., 1996). Thus, the viscoelastic properties of bitumen over a wide range of temperatures and loading are a key feature when forecasting pavement performance (Vasiljevic-Shikaleska *et al*., 2010).

Nonetheless, all pavement roads, even the best designed and constructed become deteriorated over time mainly as a result of traffic loading and weather actions (Pérez-Acebo, Bejan, *et al*., 2017; Pérez-Acebo, Mindra, *et al*., 2017). Therefore, with the aim of enhancing the bitumen performance, several blends of bitumen with a wide variety of modifiers have been analyzed (Bahia *et al*., 1998). Some examples of employed substances are sulphur (Smagulova *et al*., 2012), polyphosphoric acid (Masson, 2008), fatty acid amides (Senior-Arrieta & Córdoba-Maquilón, 2017) or polymers (Navarro *et al*., 2004). The most commonly used polymeric additives to alter bitumen properties are styrene-butadiene-styrene copolymer (SBS), styrene-butadiene rubber (SBR), ethylene vinyl acetate (EVA) and polyethylene (PE) (Bai, 2017; Moreno-Navarro *et al*., 2017; Newman, 1998; Wang *et al*., 2017; Yousefi 2003). All these modifiers can affect significantly the technical properties of resulting bitumen. For a general assessment of the modified blend, the total cost of the process must also be considered (Lu & Isacsson, 2001).

On the other hand, nowadays there is an international consensus about the necessity of a sustainable development in order to efficiently manage the limited natural resources (Pérez *et al*., 2007). In the construction industry, various waste products are reused and recycled in usual materials like concretes (Arribas *et al*., 2015) or mortars (Santamaria *et al*., 2016; Salas *et al*., 2016).

With regard to the employment of solid waste materials in asphalt pavements, a wide variety of products have been investigated, such as glass (Su & Chen 2002), steel slag, used tyres (Huang *et al*., 2007) and crushed bricks (Chen *et al*., 2001). In these cases, the recycled materials are a substitutive of the aggregates, and resulting bituminous mixtures provide sufficient characteristics to be employed in bituminous pavement roads. As previously stated, polymers have been used to enhance bitumen properties according to some requirements. On this basis, recycled polymeric materials from waste products have applied as bitumen modifiers, mainly polyethylene and ethylene vinyl acetate (García-Morales *et al*., 2004; Hinislioglu & Agar, 2004), but also polystyrene, polypropylene and reclaimed geomembranes (Al-Abdul Wahhab *et al*., 2017; García-Trave *et al*., 2018; Vila-Cortavitarte *et al*. 2018). Renewable and sustainable materials are also researched for bitumen modifiers, like natural rubber (Wen *et al*., 2017). With regard to the recycled polyurethane (PU) foam, previous researches have analyzed its employment in mortars (Junco *et al*., 2012) and in lightweight plaster (Gutierrez-Gonzalez *et al*., 2012), and even as aggregate substitute in pavement roads (Tribout & Husson, 2010). More specifically, the performance of polyurethane modified bitumen has been analyzed (Izquierdo *et al*., 2012), but in this case, the polymer was manufactured in situ, rather than providing a second use for a waste product. Similarly, Carrera *et al*. (2015) studied the use of polyurethane modified bitumen for emulsions. Once again, the polymer was not a waste product. Additionally, Padhan and Gupta (2018) proposed a new approach of in situ polymerization of monomers derived from PET waste in the body of the bitumen, which led to a homogeneous mixture of polyurethane polymer modified bitumen with successful results.

The aim of this paper is to assess the suitability of employing recycled polyurethane foam as a bitumen modifier in hot mix asphalts. Consequently, the research work not only studies the use of polyurethane waste to reduce its accumulation in landfill sites and to reduce the consumption of natural resources, facts which are of great relevance by themselves; but also studies the improvement of the bitumen and its possible applications.

Methodology

The experimental programme was performed following these steps:

1. Characterization of the polymeric polyurethane foam waste

2. Design and manufacture of modified bitumens with different percentages of recycled foam in the original bitumen

3. According to the result of modified bitumens, select the option with the greatest percentage of waste polyurethane foam providing good results.

4. Design and manufacture of the hot asphalt mix with polymer modified bitumen and compare obtained results with a standard bitumen sample.

Materials

Polyurethane foam waste

A polyurethane (PU) foam waste was employed as bitumen modifier. It is a by-product from the manufacturing of polyurethane for thermal insulation in the construction industry. The material was supplied by a company from Burgos (Spain). The waste material represents a percentage above the 5 % of the total weight of produced polyurethane in the factory. Thus, finding applications for this by-product is of great interest so as not to be disposed in landfills. The waste material is collected from the factory in the form of rigid polyurethane strips, shown in Fig. 1.

The foam was studied before their use by means of the Scanning Electron Microscopy (SEM): The chemical composition was determined by CHNS elemental analysis by means of a LECO CHNS-932 analyser with X-ray diffraction (Fig. 2). Values are shown in Table 1. The polyurethane foam was shredded to sizes between 0 and 3 mm (Fig. 3). Obtained granulometry is exposed in Fig. 4. The apparent density is 72 kg/m^3.

Figure 1. Strip of polyurethane foam.
Source: Authors

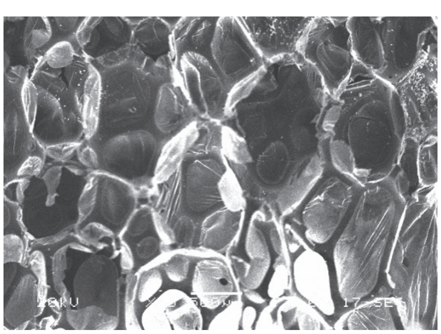

Figure 2. Electronic microscopy scan of waste polyurethane foam.
Source: Authors

Table 1. CHNS element analysis of waste polyurethane foam

Element	(%)
C	61,4
O	5,5
N	6,8
H	12,4
Others	13,9
Total	100,0

Source: Authors

Bitumen

Bitumen with a penetration grade of 50/70 was used as base material for the bitumen modification. According to Spanish regulations (Ministerio de Fomento, 2015), this type of bitumen is classified as a conventional one and fulfils the requirements listed in Table 2.

Aggregates

Crushed ophitic aggregates were employed in this study to asphalt concrete production following the experimental procedure detailed in section 2. The granulometry of the specimen fulfils the requirements for being classified as a surface layer mix, specifically AC 16 surf S, according to Spanish rules (Ministerio de Fomento, 2015) (Fig. 5). The specific weight is 2,84 g/cm3, the Los Angeles abrasion coefficient is 9,8 %, and the Polished Stone Value was 57.

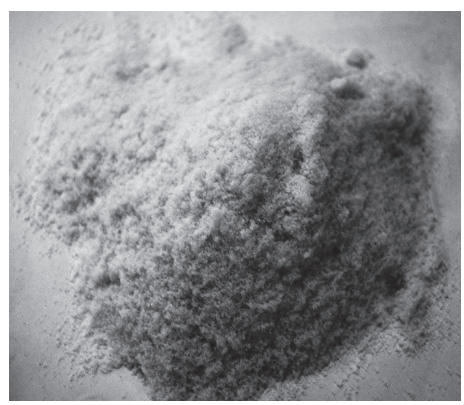

Figure 3. Appearance of shredded polyurethane foam.
Source: Authors

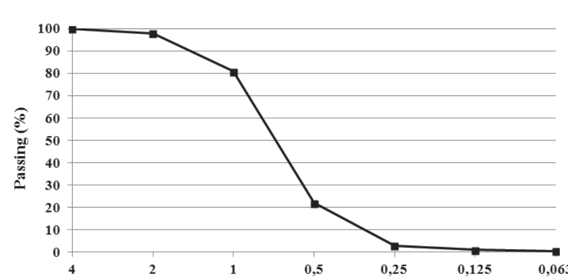

Figure 4. Granulometry of the shredded polyurethane foam.
Source: Authors

Table 2. Requirements for a bitumen B50/70

Characteristic		Standard	Units	Bitumen 50/70
Penetration at 25 °C		EN 1426	0,1 mm	50-70
Softening point		EN 1427	°C	46-54
Short-term ageing				
EN 12607-1	Mass change	EN 12607-1	%	≤ 0,5
	Retained penetration	EN 1426	%	≥ 53
	Increase in softening point	EN 1427	°C	≤ 10
Penetration index		EN 12591		
13924 EN		From -1,5 to + 0,7		
Fraas breaking point		EN 12593	°C	≤ -8
,Flash point Cleveland cup method		ISO 2592	°C	≥ 230
Solubility		EN 12592	%	≥ 99,0

Source: Orden FOM/2523/2014. Art. 542. (Ministerio de Fomento, 2015)

Results and discussion

Polymer-modified bitumen

Modified bitumen production involved the insertion of the polymer into hot bitumen (100 ºC), milling of the mixture in a high-shear mill for 1 hour and its final dissolution and homogenisation. It was performed in the factory of Firmes Alaveses in Vitoria (Spain). Various modified bitumens were manufactured from the reference bitumen by adding different amounts of polyurethane foam waste, in percentages that vary from 1 % up to 5 % of the weight of the bitumen. Table 3 shows the specimens that were elaborated and their recycled foam percentages. Substitutions of PU foam in amounts of 5 % or over were refused, as the modified blend turned into a rough material with very poor workability.

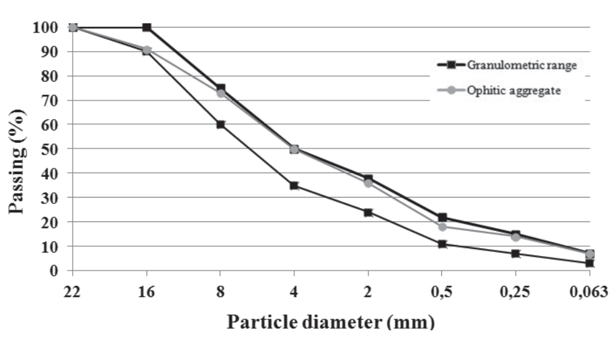

Figure 5. Granulometric curves for aggregates in AC 16 surf S and curve of the employed ophitic aggregates.
Source: Orden FOM/2523/2014. Art. 542. (Ministerio de Fomento, 2015) and authors

Table 3. Manufactured polymer modified bitumens

Sample	Characteristics	Penetration rate (10-1 mm)	Softening point (°C)
B	Reference bitumen 50/70	55	48 / 52
BM1	Bitumen modified with 1 % of recycled polyurethane foam	47	52 / 55
BM2	Bitumen modified with 2 % of recycled polyurethane foam	46	52 / 54
BM3	Bitumen modified with 3 % of recycled polyurethane foam	44	53 / 56
BM4	Bitumen modified with 4 % of recycled polyurethane foam	43	62 / 66

Source: Authors

The characterization of all the bitumens was determined by the penetration grade at 25 °C and the softening point temperature, with the ring and ball (R&B) method.

The penetration test at 25 °C is the primary method for bitumen classification in the European standardisation framework, following the standard EN 1426: 2007 (CEN, 2007a). The test involves the determination of bitumen consistency expressed as the distance that a standardised steel needle penetrates vertically into a bitumen sample at a specific temperature. The needle load is 100 g, and the loading time equals 5 seconds. The penetration unit is 0,1 mm, which corresponds to the needle penetration depth in the bitumen sample. The penetration test on the bitumens (Table 3) confirmed that, as the percentage of recycled polyurethane foam increased, the figures were lower, which indicates that the modified bitumen becomes harder as the amount of polymer was increased.

Furthermore, the softening point test was also carried out to characterize the bitumen samples. It specifies bitumen properties at high service temperatures and represents a conventional, approximate upper limit of the viscoelastic consistency. The test involves the determination of the temperature at which bitumen acquires a specific consistency. It is usually performed by the R&B method, described in the European Standard EN 1427:2007 (CEN, 2007b). Two bitumen samples placed in metal rings are heated in a controlled manner in a liquid, placed in a glass baker, with each bitumen-filled ring supporting a steel ball. The softening point is adopted as the average temperature at which both bitumen rings soften to the point that each ball, covered in bitumen and overcoming its resistance, travels a distance of 25,0 mm ± 0,4 mm. The result is expressed in °C. Results are included in Table 3 and confirmed that as the polyurethane foam is added, the softening point temperature increases.

Hot mix asphalts

As previously stated, the properties obtained with the addition of polymers in bituminous materials depend on the nature of these organic materials and their chemical structure. In general, the long chains of these compounds give the mixture greater elasticity due to the presence of very long polymeric chains. There are also short chains in the polymers, which improve adherence to aggregates. At the same time, a physical cross linking of the polymer chains occurs between the different phases that reinforce the cohesion of the mixtures, and which depends on the molecular weight and the viscosity of the added polymer.

With the mentioned aggregate sample, classified as AC 16 surf S for surface layers (Ministerio de Fomento, 2015), two series of samples were prepared (Fig. 6):

Series A: samples with original B 50/70 bitumen.

Series B: Samples with BM4 bitumen. Among the manufactured bitumens under study, bitumen BM4 was selected because it contains the largest amount of waste while maintaining acceptable properties for hardness and softening point temperature.

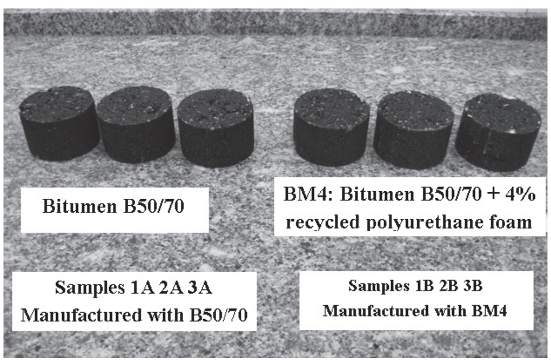

Figure 6. AC16 surf S samples manufactured with bitumen B50/70 and bitumen BM4
Source: Authors

The bituminous mixes for both series were prepared with 4,8 % of bitumen. The bulk specific density and theoretical specific density of each series was determined by the dry procedure according to European Standard EN 12697-6 (CEN, 2005), exposed in Table 4.

Table 4. Test results for series A and series B

Characteristics (Units)	HMA series A	HMA Series B	% of increase of Series B with regard to Series A
Stability (kp)	1 672	2 037	+ 21,86 %
Deformation (mm)	3,14	2,03	- 35,35 %
Voids in mix, Vm (%)	5,20	5,80	11,54 %
Void in mineral aggregate, VMA (%)	17,10	17,40	1,75 %
Aggregate voids filled with bitumen (%)	69,40	66,60	- 4,03 %
Bulk specific density of mix (kg/m^3)	2 491	2 479	- 0,48 %
Theoretical specific density of the mix (kg/m^3)	2 628	2 631	0,11 %

Source: Authors

The Marshall test was also carried out for both HMA samples. The purpose of the Marshall test is to determine the optimum content of bitumen for a specific combination of aggregates (ASTM, 2015). It is a mechanical test that consists on breaking previously prepared cylindrical test samples, heated to a temperature of 60 °C, through the application of a vertical load by means of perimetral clamp at a constant speed of deformation of 50,8 mm/min to determine their stability and deformation. Stability is measured in load units (kp), while deformability is the reduction in diameter, measured in mm, that the sample undergoes between the start of loading and the breaking point,.

The results of the Marshall tests (Table 4) show that the mixture containing the polyurethane foam has undergone a very considerable increase in stability, which entails a very significant reduction in deformation. It may suggest that these mixtures could be able to resist higher loads. Nonetheless, an increase in the percentage of air voids is also observed. It could suggest a worse performance of the mixture. However, this value is still below the limits imposed in the Spanish regulations (Ministerio de Fomento, 2015) for surface layers in any traffic category (Table 5). Consequently, the increase should not affect the ageing negatively.

Table 5. Requirements of air void in mixing

Characteristic		Heavy traffic category			
		T00 & T0	T1 & T2	T3	T4
Voids in mix (%)	Surface layer	4 – 6		3 – 6	
	Binder layer	4 – 6	4 – 7	4 – 7	4 – 7
	Base layer	4 – 7	4 – 8	4 – 8	

Source: Ministerio de Fomento (2015)

A good workability of the mixture is confirmed by the percentage of added polymer, leading to assume that on-site laying and compaction will be acceptable due to the lubricating action of the polymer.

Further research with higher percentage of waste materials should be done in order to take advantage of the possibility of using this by-product as bitumen modifier. A general characterization of the HMA is suggested to definitively validate its employment in bituminous pavement roads.

Conclusions

In this paper, preliminary results about the suitability of PU foam waste as a bitumen modifier are presented. With this aim, five samples of bitumens with different percentages of recycled polymer were produced, 1 %, 2 %, 3 %, 4 % and 5 % from an original bitumen with a penetration grade of 50/70. A greater hardness and a higher soften temperature was observed with higher percentage. The sample with

5% showed that, with that quantity, the bitumen became rough and its workability decreased severely. On the basis of a compromise between the maximum percentage of substitution and the properties of the modified bitumens, the sample with 4% (BM4) was selected to manufacture a hot bituminous mixture.

The results obtained in the Marshall test of the BM4 were compared to those obtained by hot mix asphalt with same aggregates and granulometry and the original bitumen 50/70. It was observed that polymer modified bitumen yielded considerable improvement in stability, reducing its deformability. On the other hand, a higher percentage of air voids could suggest a worse ageing performance. Nevertheless, the air voids value is still between the accepted values of Spanish rules in any traffic category.

As a conclusion, the recycled polyurethane foam waste, a by-product of construction industry, is a promising material for use as a bitumen modifier in hot asphalt mixture. Despite the low percentage that is suggested to be employed, the low density of the polyurethane foam allows to recycle a big amount of waste in bituminous mixtures. Apart from the environmental benefits of the employment of recycling waste material derived from polyurethane foam production, it also provides better characteristics to both bitumen and hot mix asphalt, achieving a better performance in bituminous pavements.

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