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Introduction of recycled polyurethane foam in mastic asphalt

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Subject review

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Introduction of recycled polyurethane foam in mastic asphalt

The aim of this paper is to analyse the suitability of recycled polyurethane (PU) foam as a binder modifier in mastic asphalts (MA). The results show that harder bitumen is achieved with an increasing percentage of PU, up to 5 %, when the workability of the sample becomes poor. A MA mix with a 4 % of PU in bitumen was manufactured. A lower indentation was observed when compared to a virgin sample, and it was established that the modified MA can be used on heavy-traffic roads. These results suggest a promising use of this waste polymer.

Key words:
polyurethane foam, mastic asphalt, bitumen modifier, polymer modified bitumen, indentation test

Pregledni rad

Miguel Ángel Salas, Heriberto Pérez-Acebo

Lijevani asfalt s dodatkom reciklirane poliuretanske pjene

U radu se analizira mogućnost upotrebe reciklirane poliuretanske pjene kao veziva/modifikatora za lijevani asfalt. Rezultati pokazuju da se tvrđi bitumen dobiva s povećanjem poliuretanske pjene do 5 % kada obradivost mješavine postaje nepovoljna. Proizvedena je mješavina lijevanog asfalta s 4 % poliuretanske pjene u bitumenu. Zabilježena je manja vrijednost dubine utiskivanja u usporedbi s uzorkom bez dodataka te je ustanovljeno da se modificirani lijevani asfalt može koristiti na cestama s teškim prometnim opterećenjem. Dobiveni rezultati pokazuju da postoje dobri izgledi za širu primjenu ovog otpadnog polimera.

Ključne riječi:
lijevani asfalt, poliuretanska pjena, modifikator bitumena, polimerom modificirani bitumen

Übersichtsarbeit

Miguel Ángel Salas, Heriberto Pérez-Acebo

Gussasphalt mit Zusatz von wiederverwertetem Polyurethanschaum

In der Abhandlung analysiert man die Möglichkeit der Verwendung von wiederverwertetem Polyurethanschaum als Bindemittel/Modifizierer für Gussasphalt. Die Ergebnisse zeigen, dass man Hartbitumen durch eine Erhöhung des Polyurethanschaums bis 5% erhält, wenn die Verarbeitbarkeit der Mischung schlecht wird. Hergestellt wurde eine Gussasphaltmischung mit 4% Polyurethanschaum im Bitumen. Verzeichnet wurde ein geringerer Wert der Einpresstiefe im Vergleich zur Probe ohne Zusatz, und es wurde festgestellt, dass der modifizierte Gussasphalt bei Straßen mit einer schweren Verkehrsbelastung verwendet werden kann. Die erhaltenen Ergebnisse zeigen, dass es gute Aussichten für eine breitere Verwendung dieses Abfallpolymers gibt.

Schlüsselwörter:
Gussasphalt, Polyurethanschaum, Bitumenmodifizierer, polymermodifiziertes Bitumen

1. Introduction

The mastic asphalt (MA) can be defined as a dense mixture consisting of coarse aggregate, and/or sand and/or fine aggregates, and/or filler and bitumen, which may contain additives [1]. It is different from other asphalt mix types due to its composition, application and load transfer [1]. Firstly, with regard to its composition, the principal MA components are the bituminous binder, with the content ranging from 6.0 M% to 9.5 M% (percentage by mass) depending on its use [2] (and a usual value around 8 M% [1]); and filler (material ≤ 0.063 mm) with up to 30 M% [2]. Hence, up to 40 M% of the mixture is regarded as mastic (binder + filler), while the remaining portion of the mixture is formed of coarse aggregates. These figures are different from those related to asphalt concrete (AC), where mastic is represented with only 10-15 M% [1]. Secondly, MA transfers loads mainly by means of stiff mastic and not by means of the coarse aggregate skeleton, as in AC. Lastly, MA has a wide field of application. Main uses of MA include bridge decks, waterproofing on flat roofs, and pavement surface layers in city centres [3-5]. Since 1960, MA mixes have been deployed on bridge decks in Europe and Japan because of their superior waterproofing capabilities and higher flexural resistance compared to other materials [3]. Moreover, the use of MA could avoid the compaction vibration of usual AC, which could be harmful for historic buildings in city centres. It is also employed on sidewalks or bike lanes, where layer thicknesses of 30-50 mm are required. Other applications of MA include flooring in buildings and factories, rooftop car parks, hydraulic constructions, and tanking [1].

Mastic asphalt is mixed and manufactured at very high temperatures ranging from 220 °C to 240 °C. Because of its low viscosity, MA does not require compaction when applied in the field: it is merely poured and self-compacted [1]. MA is placed at high temperatures either using screed pavers or manually [6]. Furthermore, air voids are almost non-existent in MA.

Since all paved roads, including even the best designed and constructed roadways, deteriorate over time due to traffic load, material ageing, or environmental effects [7-10], bitumen additives have been widely employed to enhance bitumen performance. Several modifiers have been analysed in this respect. Examples of substances used for that purpose are sulphur [11], polyphosphoric acid [12], mineral acids [13], waxes [14, 15], or polymers. Among polymers, styrene-butadiene-styrene (SBS), styrene-butadiene rubber (SBR), ethylene vinyl acetate (EVA), and polyethylene (PE), are the most widely applied [16-18]. Similarly, as mastic is the key load transferring component, additives - mainly waxes, rubbers, fibres, pigments and polymers - are added to MA [2, 6]. On the other hand, an international consensus has been reached over the last decades about the need for a more sustainable development and a more efficient management of natural resources [19]. It has been widely reported that industrial countries are producing a substantial amount of waste. Generally, materials are extracted, directly used or processed into final products, which are then consumed and finally placed in landfills or incinerated [20]. The European Union is taking steps to improve resource efficiency and reduce environmental and climate impacts by promoting the reuse and recycling of waste materials [21]. Some waste materials are recycled back into the same product. Nonetheless, despite progress of such recycling techniques, some quality requirements are not fulfilled by recycled materials and, hence, these materials are refused. This means that construction industry has a great opportunity to transform waste materials into raw materials [20]. References on this issue can be found in relevant literature [22, 23].

Concerning road pavements, various waste materials and products have been incorporated, and successful results have been obtained. Two pavement recycling goals can be differentiated: preserve resources and improve material properties [20]. Waste materials that are most commonly used in current practice are commented below.

Crushed stone from waste concrete is the main material generated in construction and demolition works. It has dominantly been used as coarse aggregate in base and sub-base layers [24]. Successful results have also been achieved by adding this material to asphalt mixes [25]. Reclaimed asphalt pavement (RAP) is a very commonly reused material, and it is mainly employed in subbase layers and bituminous layers [26-29]. AC with high proportions of RAP can exhibit similar mechanical performance [30]. The employment of glass in road construction has also been investigated, but the main difficulty lies in particle morphology [31, 32]. Scrap tires have been thoroughly examined to determine their usability in asphalt mixtures. The most interesting developments have been identified in mixtures modified with crumb rubber, which can be added to the bitumen or to the mixture itself [33-35]. Ceramics also represent an alternative source of aggregate. They were initially employed in sub-bases or concrete pavements [36, 37], but research about their use in Stone Mastic Asphalt has also been published [38]. Steel slag has been largely used in road construction in Europe [39].

Thanks to extensive research about polymer modified bitumens, many studies are currently available about the use of waste polymers in roads [40]. Some of the most commonly employed recycled polymers are discussed. Recycled polyethylene terephthalate (PET) has been principally used in road construction as aggregate substitute [41, 42], and as bitumen modifier and anti-stripping agent if chemically

processed [43, 44]. In Stone Matrix Asphalt, recycled PET has been found to increase resistance to permanent deformation and stiffness [45, 46]. By means of PE modified bitumens, improvements have been achieved in Marshal Stability [47], rutting resistance [48], penetration softening point and ductility [49]. Bitumen properties in the low and high temperature ranges have been improved by adding EVA [50]. However, crosslinking agents and catalysts must be carefully selected to enable EVA addition [51]. Polyurethane (PU) can be found in several applications: food cold chain, in furniture, mattresses, shoes, cars, and in thermal insulation of buildings. Generally, PU ends up in landfills or incinerators, and only a small portion (10 %) is recycled. Together with a large quantity of excavation, construction and demolition waste, PU represents around 30 % of all waste material in the European Union [20]. Despite the incompatibility of PU with bitumen, which results in an instable system [52], this problem has been overcome either by means of plasticizers or by pre-treating bitumen with modified clay, maleic anhydride, or dibasic acids. Waterproofing coating/sealing compounds were improved adding a PU pre-polymer [53]. Carrera et al. [54] used PU as a reactive polymer which was added to bitumen compounds for obtaining PU modified bitumen. In this case, the bitumen modification occurs due to reaction of isocyanate groups (-N = C = O) of the pre-polymer with functional groups, which contain active hydrogen atoms, generally –OH, normally present in asphaltene micelles. The influence of the molecular weight and isocyanate content on the rheology of PU bitumens has also been studied [55].
More specifically, concerning the polyurethane foam waste, previous research focused on its use in mortars [56, 57] and lightweight plasters [58]. In road construction, Izquierdo et al. [59] analyzed the employment of PU for bituminous foam because of the ability of the isocyanate compounds to be foamed, the aim being to combine waterproofing characteristics of bitumen with thermal and acoustic insulation properties of sprayed PU foams to form a new product. It was evaluated as effective, but the polymer was manufactured in situ, rather than providing a second use for a waste product.
The aim of this paper is to examine suitability of the use of waste polyurethane foam as bitumen modifier in mastic asphalts. Therefore, apart from studying possibility of using the polyurethane foam waste in road construction thus avoiding its accumulation in landfills and reducing consumption of limited natural resources - facts which are of great relevance by themselves - it also investigates possibilities for improvement of mastic asphalt properties and its applications.

2. Methodology

The experimental program consisted of the following steps. Initially, the polymeric polyurethane foam waste was characterized, including its origin, form in which it was supplied, chemical composition, apparent density, and grading after the shredding process. Secondly, modified bitumens with different weight percentages of recycled foam in original bitumen were designed and manufactured. The penetration and softening point test (by means of the ring and ball method) were conducted to characterize each specimen. Then, the option with the highest quantity of waste polyurethane providing suitable results was selected according to the results for previous modified bitumens. In the final step, mastic asphalt with polymer modified bitumen was designed and manufactured, and the results obtained during the indentation test were compared to the MA mixture with the standard bitumen sample.

3. Materials

3.1. Polyurethane foam waste

A thermostable polyurethane waste was used as bitumen modifier. It is a by-product obtained during manufacture of polyurethane used for thermal insulation in construction industry. The polyurethane foam was obtained from a company from Burgos (Spain), where this waste material accounts for more than 5 % of the total weight of the polyurethane produced in the factory. Hence, the possibility of finding an alternative application for this by-product is very interesting as a means to avoid disposal to landfill sites. The waste material was supplied by the company in form of rigid polyurethane strips, as shown in Figure 1.

Figure 1. Waste strip of polyurethane foam supplied by the factory

The polymer was examined using the Scanning Electron Microscopy (SEM) before being deployed. The chemical composition was determined by CHNS elemental analysis using the LECO CHNS-932 analyser and X-ray diffraction (Figure 2). The results are shown in Table 1.

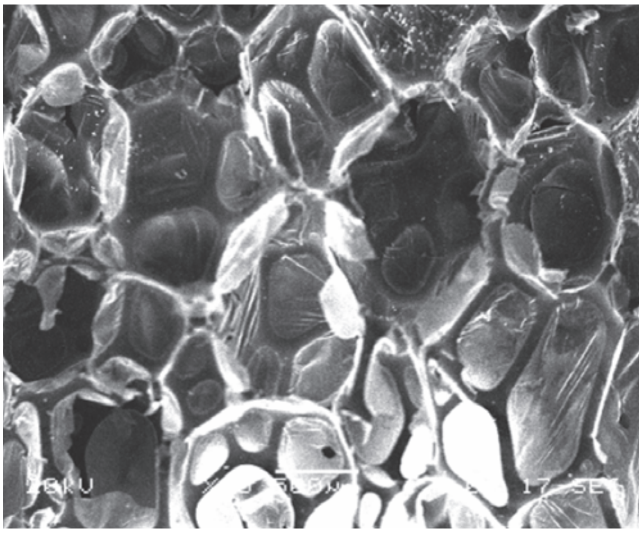

Figure 2. Electronic microscopy scanning of waste polyurethane foam

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**

All polyurethanes are organic compounds based on the reaction of two different chemicals: a polyol (with – OH terminal groups) and a di-isocyanate (with – N = C = O terminal groups), which react due to a catalyst that is added in small quantities to activate the polymerization reaction. Therefore, the chemical composition values - exhibiting high levels of carbon, oxygen, nitrogen and hydrogen – are correct. Other components were found in very low quantities and, hence, it was concluded that they are residual components, belonging to the catalyst.

To be suitable for use as bitumen modifier, the polyurethane foam strips must be shredded. This was carried out by means of a waste shredding machine, Retsch SM 100, which allows shredding to different sizes. Sizes between 0 and 3 mm were selected. Material remaining after the shredding process is shown in Figure 3. The resulting grading is presented in Figure 4. The apparent density of the shredded foam is 72 kg/m^3. Finally, the polyurethane foam was not catalogued as hazardous waste [60].

Figure 3. Appearance of shredded polyurethane foam

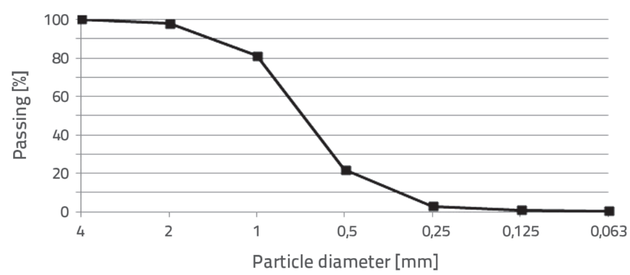

Figure 4. Grain size distribution of shredded polyurethane foam

3.2. Bitumen

For mastic asphalt manufacturing, a bitumen with the penetration grade of 35/50 was selected as base material for modifications. It was supplied by the company Balgorza sna,

Table 2. Requirements for bitumen B35/50 [61]

Characteristic		Standard	Units	Bitumen 35/50
Penetration at 25 °C		EN 1426	0.1 mm	35-50
Softening point		EN 1427	°C	50-58
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	≤ 11
Penetration index		EN 12591 EN 13924		from -1.5 to + 0.7
Fraas breaking point		EN 12593	°C	≤ -5
Flash point, Cleveland cup method		ISO 2592	°C	≥ 240
Solubility		EN 12592	%	≥ 99.0

based in Vitoria-Gasteiz (Spain). This type of bitumen fulfils requirements specified in Spanish regulations [61], as shown in Table 2.

3.3. Aggregates

Aggregates were supplied from the quarry Ofitas de San Felices located in Haro (Spain). The maximum nominal aggregate size of 11 mm (MA 11) was adopted for this study. The aggregates are of ophitic origin and are totally crushed. The specific weight is 2.84 g/cm^3, the Los Angeles abrasion coefficient is 9.8 %, and the Polished Stone Value amounts to 57. Powdered limestone was used as filler component. Additional mechanical property requirements for use in MA [2] were fulfilled.

4. Results and discussion

4.1. Polymer modified bitumen

The procedure was carried out at the factory of Firmes Alaveses in Vitoria-Gasteiz (Spain). Various modified bitumens were prepared by adding different quantities of polyurethane waste to the reference bitumen. The added percentages ranged from 1 M% (percentage by mass) to 5 M%. The addition of polyurethane foam in the amounts of 5 M% or more without a catalyst was discarded, as the modified bitumen turned in such cases into a rough material with very poor workability. This handicap could be avoided by adding reactive or pre-polymer, as was done by other authors [53, 54].

The characterization of the reference bitumen and the manufactured modified bitumen was performed by means of penetration at 25 °C and the softening point temperature, using the ring and ball (R&B) method.

The penetration test is the primary test for bitumen classification in the European standardization framework, as established in EN 1426:2007 [62]. As a general rule, lower penetration denotes harder bitumen. Penetration test results are presented in Table 3. As can be seen, lower values are obtained if a higher percentage of recycled PU foam is added. This means that the modified bitumen becomes harder with the addition of polymer. The softening point test was also conducted for reference and modified bitumen specimens. It was carried out by the ring and ball method (R&B), defined in the European Standard EN 1427:2007 [63]. It gives information about bitumen performance at high service temperatures, and it is said to represent a conventional, approximate upper limit of viscoelastic consistency. The test determines the temperature at which bitumen acquires a specific consistency. The results obtained are given in Table 3. In concordance with previous penetration test results, a higher quantity of polyurethane foam results in an increase in softening point temperature.

As can be seen, with the upper limit of 5 M%, the more waste PU foam is added to the bitumen, the harder it becomes, which results in poor workability. Since the values obtained for penetration and softening points are between usual limits for this kind of binders, the percentage of 4 M% was selected for mastic asphalt production and evaluation because of its higher hardness and higher percentage of waste material.

4.2. Mastic asphalt

Two mastic asphalt specimens were manufactured. The first one, called N11 Normal, was produced without any polymer and

Table 3. Manufactured polymer modified bitumens

Sample	Characteristics	Penetration [10^{-1} mm]	Softening point [°C]
B	Reference bitumen 35/50	41	53.5
BM1	Bitumen modified with 1 M% of recycled polyurethane foam	34	56.5
BM2	Bitumen modified with 2 M% of recycled polyurethane foam	33	57.5
BM3	Bitumen modified with 3 M% of recycled polyurethane foam	31	58
BM4	Bitumen modified with 4 M% of recycled polyurethane foam	29	65.5

Table 4. Grading employed in mastic asphalt samples (N11 Normal and N11 PUR)

Sieve [mm]	% passing							
	11.2	8	5.6	4	2	0.5	0.25	0.063
N11 Normal	100	100	100	86	57	41.3	36	28.1
N11 PUR	100	100	100	81	56	41	36	27.4

Table 5. Percentage composition of manufactured MA specimens

Sample	Cooking temperature [°C]	Binder/aggregate [%]	Filler/ bitumen [%]	Density [g/cm^3]
N11 Normal	230	8.43	3.30	2419.70
N11 PUR	230	8.70	3.10	2403.80

Table 6. Indentation test for mastic asphalts [65]

	W test	A test	B test	C test	D test
Temperatur [°C]	25	25	40	40	22
Rod area	31.7 mm^2	5 cm^2	5 cm^2	1 cm^2	1 cm^2
Applied load [kg]	31.7	52.5	52.5	52.5	52.5
Load application time	1 min 10 s	6 min	31 min	31 min	300 min
Measure period	from 10 s to 70 s	from 1 min to 6 min	from 1 to 31 min	from 1 to 31 min	from 1 to 300 min

Table 7. Indentation test used for each mastic asphalt application and required indentation test values

Application	Reference name	Applied indentation test	Usual layer thickness [mm]	Required values [10^{-1} mm]
Pedestrian areas	AP	B test	20 – 25	20 ≤ I ≤ 50
Roads	AVL	B test	25 – 30	10 ≤ I ≤ 30
Heavy traffic roads	AVP	B test	30 – 40	5 ≤ I ≤ 15
Acoustic isolation of apartment floors	AIP	C test		1 ≤ I ≤ 12
Indoor industrial floors	AIC	C test	25 – 30	10 ≤ I ≤ 30
Heavy indoor industrial floors	AIP	C test	25 – 30	1 ≤ I ≤ 12
Anti-acid floors		C test		10 ≤ I ≤ 30
Sports areas		B test		20 ≤ I ≤ 50

was used as reference MA. The second one, called N11 PUR, was manufactured with 4 M% of recycled PU foam, based on the values of the polyurethane foam waste modified bitumen tests. The specimens were produced in accordance with the European Standard EN 13108-6 [2]. Aggregate gradings, selected for both specimens in accordance with EN 13108-6 [2], are shown in Table 4.

The percentages of each component in both samples N11 Normal and N11 PUR are listed in Table 5.

The indentation test [64], together with Hardness Number, has been established in European standards on mastic asphalts as a reference test for representing ability of a material to resist permanent deformation. According to Spanish standards on MA application [65], there are five indentation test possibilities relating to the use of mastic asphalt (Table 6). Table 7 shows the type of indentation test used for each mastic asphalt application and required indentation test values.

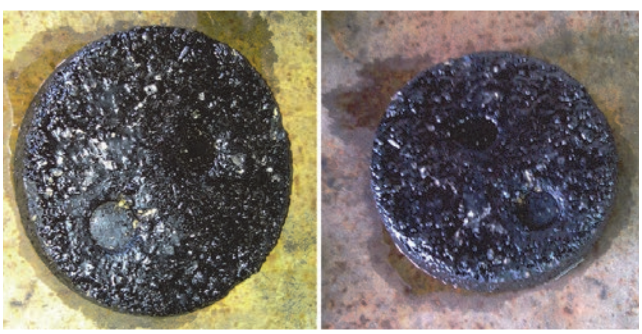

Figure 5. Mastic asphalt samples after indentation test, N11 Normal (left), N11 PUR (right).

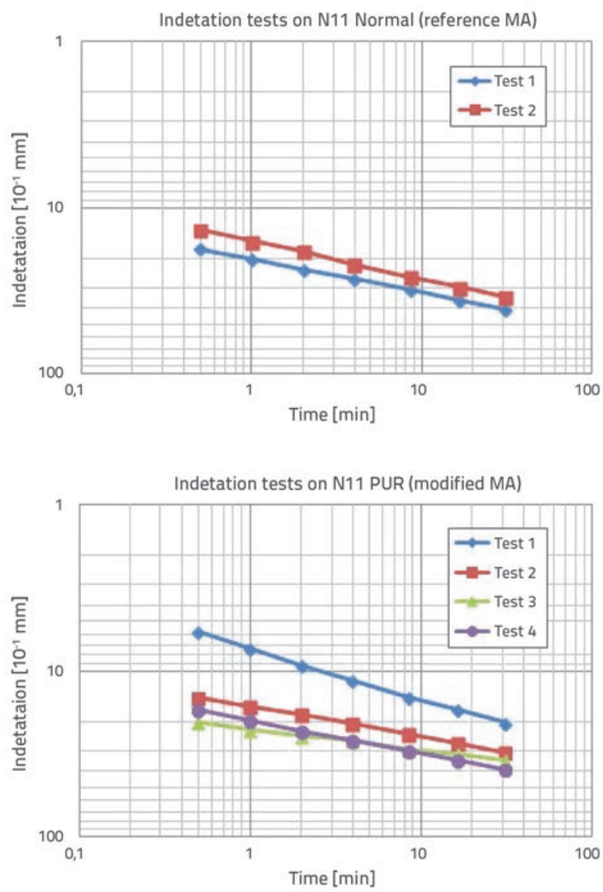

Figure 6. Indentation test results for N11 PUR: reference sample (up) and polymer modified sample (down)

Table 8. Summary of indentation test results

Specimen		Indentation depth in 1 min [10^{-1} mm]	Indentation depth in 31 min [10^{-1} mm]	Indentation value [10^{-1} mm]
N11 Normal	Test 1	20.2	41.0	20.80
	Test 2	15.8	34.3	18.50
	Average			**19.65**
N11 PUR	Test 1	7.3	20.4	13.1
	Test 2	16.2	30.8	14.6
	Test 3	22.3	34.6	12.3
	Test 4	19.9	38.9	19.0
	Average			**14.75**

Mastic asphalt for use on roads was investigated in this research, and the indentation test B was carried out. Two tests were conducted on specimen N11 Normal and four tests on specimen N11 PUR (Figure 5). Results of these tests are shown in Figure 6.

As can be seen, indentation test results obtained for specimen N11 Normal are more homogeneous than the ones for N11 PUR. Nevertheless, the final test results, summarized in Table 8, show that a lower indentation value was achieved on modified MA.

According to Table 7, mastic asphalt N11 PUR could be employed in heavy traffic due to improvement in mechanical properties of the mixture. This means an improvement was noted compared to the initial mastic asphalt, N11 Normal, with no modifications. An important reduction of indentation value, from 19.65 to 14.75 (24.9 %), was obtained due to introduction of the PU foam waste, which permits use of the new MA mixture on heavy traffic roads. Additionally, a little variation of the density was observed (Table 5).

Therefore, the possibility of using waste polyurethane foam was validated by means of these tests. Initially, the bitumen with higher quantity of foam was selected. It was the harder modified binder as the penetration test showed, with the highest softening point. When manufacturing a mastic asphalt with this percentage, the higher hardness is reflected in lower values of indentation tests, allowing deployment of this material on roads with heavy traffic. The new material shows an improved load related performance, and so the two main objectives of the research were accomplished: contribution to waste material reduction and, at the same time, enhancement of material characteristics.

5. Conclusions

In this preliminary study, some laboratory tests were performed to evaluate feasibility of using polyurethane foam waste in mastic asphalt. A harder bitumen was obtained as the amount of PU was increased from 1 M% to 4 M% (weight by mass). The specimens with 5 M% of PU or more were discarded due to their poor workability, which would cause problems during the manufacture and placing of mastic asphalt mixtures. The addition of polymer is suggested during the modified bitumen manufacturing to increase the polyurethane foam waste percentage over 5 M% with improved workability.

The softening point test, carried out by the ring and ball method, revealed an increase in softening temperature with an increase in the quantity of PU in the mixture.

Based on these results, a mastic asphalt mixture with a 4 M% of polyurethane foam waste was manufactured and compared with a standard MA without additions in the bitumen. Mechanical properties of the mastic asphalt with PU foam waste modified bitumen were improved, as reflected in the indentation test. While the reference MA was prepared for use in normal traffic road surfaces, the values obtained in the indentation test make the modified MA mixture suitable for heavy traffic road surface. An improvement in indentation values of almost 25 % was achieved, with no variations in density.

It must be noted that these conclusions are based on a limited number of specimens and tests. Further research is needed to introduce the possibility of using polyurethane foam waste as binder modifier in mastic asphalt regulations. As a conclusion of this introductory approach, it can be stated that the influence of PU foam waste in mastic asphalt mixtures is positive.

In brief, it can be concluded that two main objectives of the research were achieved. On one hand, an enhanced mastic asphalt that can be used on heavy traffic roads was produced. On the other hand, the new material utilizes waste material that would otherwise be disposed on landfills. Consequently, a better and more sustainable material is proposed.

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