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The influence of recycled plastics added via the dry process on the properties of bitumen and asphalt mixtures

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A R T L C L E I N F O

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ABSTRACT

Nowadays, the use of recycled plastics is gaining momentum, especially after the 2017 plastics crisis, and the asphalt pavement industry has positioned itself as a leader due to its long history of using recycled materials. However, regardless of the potential interest in using recycled plastics in road infrastructure, there is still a lack of knowledge on how to make their incorporation into asphalt mixtures a daily practise. For this reason, a study was conducted to evaluate the influence of five patented recycled plastics: on bitumen properties, on the affinity between aggregate and bitumen, on the resistance of five asphalt mixtures (AC14 surf) to permanent deformation and moisture, and on their skid resistance. The recycled plastics used consist mainly of PE (low and high density) and PP and were added via the dry process due to the simplicity and low cost of the process. Results show that recycled plastics influence bitumen properties, but do not improve ageing resistance. In addition their use tends to improve the affinity between aggregate and bitumen and influence the behaviour of asphalt mixtures: specimen porosity is increased, Marshall properties generally remain similar or increase slightly, water sensitivity is not significantly affected or slightly improved, and surface properties remain similar for macrotexture and exhibit higher slip/skid resistance. No significant relationship was found between bitumen properties and other properties studied. Regardless of the type, form, plastic content, replacement, or increase in bitumen content used in this study, recycled plastics are an alternative t60 other techniques for pavement construction/ rehabilitation.

1. Introduction

Plastic is one of the most widely used materials in the world because it is technically advanced, lightweight and cheap. The amount of plastic used has steadily increased over the past 30 years, reaching over 390 million tonnes in 2021, with a projected exponential increase to 600 million tonnes by 2025 [21,22]. Plastic waste represents a significant and growing environmental challenge, including industrial plastics, plastic bags, food packaging, and others [2,5,6]. Since the 1950s, nearly 50% of all plastics end up in landfills or in the wild, and only 9% of the plastics used have been adequately recycled. It is estimated that 4 to 12 million tonnes of plastic waste end up in the oceans [2]. In response to this growing concern, there has been increased interest in using this waste in other applications, particularly the incorporation of recycled and processed plastic waste into building materials as a means of promoting the circular economy [8,16,30,32]. Plastics are synthetic materials derived from refined petroleum and are characterised by a high softening temperature and high resistance to ultraviolet (UV) radiation [9,27]. The above properties bring a number of advantages when plastics are to be used in asphalt mixtures, as an additive to improve the properties of bitumen or to partially replace the bituminous binder. However, they can also bring disadvantages, as their residues remain in the environment for hundreds of years, posing an increasing environmental challenge. In addition, the toxic chemicals contained in many plastics are bioaccumulative, thus posing a risk to the health and safety of the food chain, including humans [27].

The two largest sources of plastic waste are plastics from plastic bottles and single-use bags. Plastic bags are made from high-density polyethylene (HDPE) and plastic bottles are made from polyethylene terephthalate (PET). HDPE has a melting temperature of up to about

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150 °C (lower than the temperature used to produce asphalt mixtures) and PET has a melting temperature of about 250 °C. Therefore, when adding plastics to asphalt mixtures, a distinction can be made between plastics with a low melting temperature, which are close to the temperatures used for the production and storage of the asphalt mixtures or bitumens, respectively, such as plastics that can replace or modify the bitumens, and those with a high melting temperature, which can replace part of the aggregates that compose the asphalt mixture.

There are two main types of processes by which recycled plastics can be incorporated into asphalt mixtures: the wet process and the dry process. In the wet process, the recycled plastic is added directly into the bitumen, which modifies the bitumen's properties and/or replaces it partially. This process requires mechanical mixing with a high-shear to obtain a homogeneous modified binder blend (bitumen + recycled plastic). Only recycled plastics with a low melting point, such as linear low-density polyethylene (LLDPE), low-density polyethylene (LDPE), and high-density polyethylene (HDPE), are suitable for this process, and are reported in the literature in dosages ranging from 2 to 8% of the bitumen mass [11,25]. Several advantages are reported in the literature, such as the extension of the durability of the asphalt layer, an improvement in the performance of the asphalt mixture, and the possibility of controlling the properties of the plastic modified bitumen [1, 12,26]. These previous improvements related to asphalt mixtures result from an increase in adhesion, an increase in complex modulus at high temperatures, an increase in cohesion and elasticity, a decrease in susceptibility to thermal variations, an increase in durability, and resistance to fatigue and permanent deformation ([11,14,15,17]; Muzaffar [19, 24]).

In the dry process, the recycled plastics are added directly to the hot aggregates during the production of the asphalt mixtures. The added recycled plastics can present three functions: aggregate replacement, mixture modifiers, bitumen modifiers, or a combination thereof [29,31]. The aggregate replacement is commonly used with recycled plastics with a high melting point (>200 °C), that is higher than the typical production temperature of asphalt mixtures, such as polyethylene terephthalate (PET), polystyrene (PS), polycarbonate (PC), and others, with the exception of polyvinyl chloride (PVC) due to chloride emissions. Those plastics can also have the function of mixtures modification as the interactions between bitumen and plastic aggregates are different from the interactions with aggregates. These interactions depend on the physical and mechanical properties of the plastics. The recycled plastics with a melting point similar to or lower than the production mixture temperature, e.g., PP, LLDPE, LDPE, and HDPE, are used for mixture modification through a partial bitumen modification in a dry process [9, 12,18,23,28,29]. It has been identified that the addition of recycled plastics by dry process is more advantageous, as it allows the incorporation of higher percentages of plastics compared to the wet process and does not require significant changes in the mixture production process. Depending on the type of plastics used and their characteristics they can perform different functions, namely, sealing the aggregates, resulting in plastic-coated aggregates, preventing water absorption and improving their physical and surface characteristics; changing the bitumen properties, as part of the plastic can melt and blend with the bitumen, and partially changing the aggregate fraction [1,18]. The changes in the bitumen and aggregate properties can affect the stability, water sensitivity, permanent deformation, and fatigue cracking of the asphalt mixtures [4,7,13,20]. In addition, the changes in bitumen matrix can prevent bitumen ageing [1,18].

Despite these efforts, most research studies focus on the addition of recycled plastics via the wet process and the use of polyethylene, PE (LDPE, HDPE, and LLDPE), and to a lesser extent the dry process and recycled plastics such as polypropylene, PP [30]. However, the dry process is a simpler and more feasible technical process that is compatible with the existing technology of asphalt mixing plants in Portugal. Therefore, this study aims to contribute to a better understanding of the influence of recycled plastics (PP as well as PE) added via

the dry process on bitumen properties and their relationship with other properties commonly investigated in studies on the use of recycled plastics in asphalt mixtures, such as Marshall properties, resistance to permanent deformation, less common properties such as affinity between aggregate and bitumen, water sensitivity, skid resistance of asphalt mixtures, and dispersion of recycled plastics in the asphalt mixture with recycled plastics added via the dry process.

2. Materials and methodology

2.1. Materials

The Portuguese highway network covers more than 3000 km and has special conditions in terms of high volume of heavy traffic and different climatic zones from the south to the north of Portugal. The existing pavement structures of the highway network have an overall thickness that allows major rehabilitation works to be concentrated mainly on the surface layer. Therefore, an AC 14 surf (gap graded) was selected in this study. This asphalt mixture is used as a surface layer in much of the highway network due to its high performance and durability. The grading curve of this type of asphalt mix more or less resembles a Stone Mastic Asphalt (SMA) with low air voids and good macrotexture, but without the need for stabilizer additives or high filler and bitumen contents (5.0-5.5% in the latter case). Fig. 1 shows the aggregate grading envelope and the grading curve of the AC 14 surf used in this study compared to the aggregate grading envelope of a SMA 14 surf specified for airfields in the United Kingdom, UK [3]. Conventional granodiorites (coarse and fine) and limestone (filler) were used as aggregates.

Polymer modified bitumen, classified as PMB 45/80-65, was used as a reference (designated as B2-REF) plus a 50/70 (B1), as shown in Table 1 for a combination of five patented recycled plastic additives (P1, P2, P3, P4, and P5, for confidentiality reasons) available on the national and international market (see Fig. 2).

From Fig. 2, it can be seen that the recycled plastic additives used have different forms, from plastic chips composed of different types of recycled plastic (P2 and P3, post-consumer plastics) to industrially processed pellets composed of different types of recycled plastic (P1, post-consumer plastics; P4 and P5, pre and post-consumer plastics). The recycled plastics used in the additives are mainly low density polyethylene (LDPE) and high density polyethylene (HDPE), and polypropylene (PP) in the case of P1 (part of the additive) and P5 (the entire additive).

Fig. 3 shows the general approach to the development of this study, including the selection of recycled plastics.

According to Fig. 3, a survey (not an enquiry/questionnaire) was conducted in the first phase to identify and select national and international manufacturers with eligible additives (recycled plastics) that can be added via the dry process (regardless of the manufacturer's industrial production process). The additives P4 and P5 are suitable for both processes (wet and dry process), but in this study only the dry process was used.

In a second phase, the content of recycled plastics and their function in the asphalt mix (aggregate replacement, mixture modifiers, bitumen modifiers, or any combination thereof) were discussed with additive manufacturers, the main objective being to ensure that the use of recycled plastics contributes to the same or improved performance compared to the reference mix AC 14 surf with polymer modified bitumen (PMB 45/80-65). Therefore, in accordance with the best practice of manufacturers:

• P1 recycled plastic additive was selected for use with a bitumen 50/ 70, B1 (to avoid highly modified bitumen). The remaining plastic additives (P2, P3, P4 and P5) were used with a polymer modified bitumen (PMB 45/80-65, B2-REF).

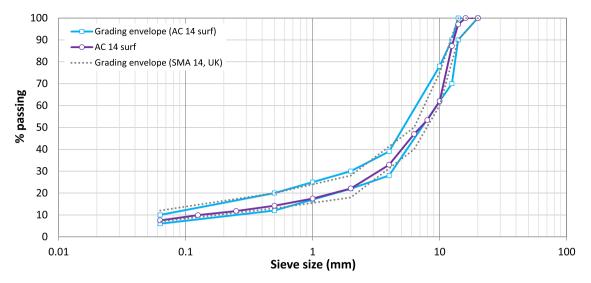


Fig. 1. Asphalt mixture aggregate grading curve and envelope.

Table 1

Properties of base bitumen.

Properties ¹⁾		Bitumen o	lassification
		50/70, B1	PMB 45/80–65, B2- REF
Penetration, 25	5 °C (0.1 mm)	60	48
Softening temp	erature (°C)	51.6	72.6
Elastic recovery	y, 25 °C (%	-	88
Cohesion energy	y, 10 °C (J/cm ²)	-	4.5
RTFOT, 163 °C	Penetration, 25 °C (0.1 mm)	-	36
	Softening temperature, (°C)	-	75.6
	Mass loss (%)	-	0.4

Abbreviations: RTFOT - rolling thin film oven test; PMB - polymer modified bitumen.

1) Penetration, according to EN 1426; Softening temperature, according to EN 1427; Elastic recovery, according to EN 13398; Cohesion energy, according to EN 13589; RTFOT, according to EN 12607–1.

• P1, P2 and P3 were added as partial replacement of bitumen content. P4 and P5 were added in addition to the bitumen content (in all cases considering the optimum bitumen content of the reference asphalt mixture).

Table 2 summarises the composition of the test specimens used in the study.

Following the selection of materials, the third, fourth and fifth phases (Fig. 3) included the addition of recycled plastics into the bitumen/ asphalt mixture, the experimental test programme to investigate the influence of the recycled plastics on the behaviour of the bitumen, on the affinity between aggregate and bitumen, on the Marshall properties and on the behaviour of the asphalt mixture (mechanical and surface properties), as described below.

2.2. The addition of recycled plastics into bitumen/asphalt mixture

The following describes the addition of recycled plastics into bitumen/asphalt mixture (third phase of the study). As a first step, the addition of recycled plastics into bitumen was evaluated to further investigate the effects of recycled plastics on bitumen behaviour (fourth phase). Initially, two methods of adding recycled plastics to bitumen

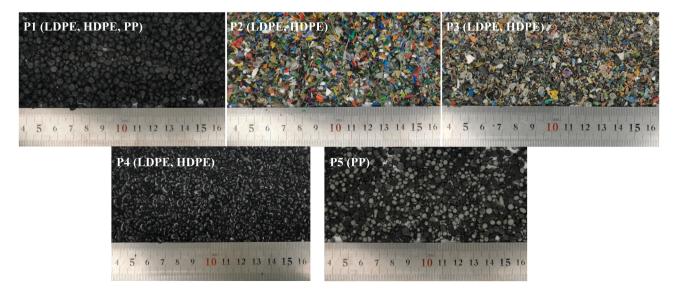


Fig. 2. Recycled plastics used in the study.

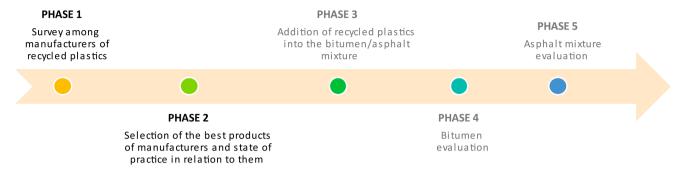


Fig. 3. Approach used in the study including the selection of recycled plastics.

Table 2	
Composition of the test specimens.	

ID	Test specimens' ty	Test specimens' type		Bitumen	Bitumen	Recycled	Main recycled	Plastic
	Bitumen or loose material ¹⁾	Compacted material ²⁾		classification	content (%)	plastic forms	plastics in the additive	content (%)
B1		_	Granodiorites	50/70	5.0	-	_	0.0
B2-REF			(course and fine)	PMB 45/80-65		-	-	0.0
B1P1			Limestone (filler)	50/70		Pellets	LDPE, HDPE, PP	5.0 ⁴⁾
B2P2				PMB 45/80-65		Plastic chips	LDPE, HDPE	6.0 ⁴⁾
B2P3						-		
B2P4	-					Pellets		$2.0^{(5)}$
B2P5							PP	

Abbreviations: LDPE - low density polyethylene; HDPE - high density polyethylene; PP - polypropylene; PMB - polymer modified bitumen. 1) Test specimens used to evaluate bitumen properties or the affinity between aggregate and bitumen.

2) Test specimens of AC 14 surf to evaluate Marshall properties, water sensitivity, permanent deformation, and skid resistance.

3) To evaluate affinity between aggregate and bitumen, Marshall properties, water sensitivity, permanent deformation and skid resistance.

4) Partial replacement relative to bitumen content.

5) Increment relative to bitumen content.

were considered (Table 3):

- Manual mixing of the bitumen with recycled plastic additives using a smooth rod (Fig. 4) for about one minute to simulate reduced interaction between the bitumen and the plastics during the production in the asphalt mixing plant.
- Mechanical mixing of the bitumen with the recycled plastic additives with a mixer at 600 rpm (Fig. 4) for five minutes (P1), sixty minutes (P2, P3), and thirty minutes (P4, P5). The differences in the time required result from the information provided by the additive manufacturers and, in the case of additive P1, from the heavily modification observed during manual mixing with the bitumen.

The bitumen was heated to the mixing temperature for asphalt mixtures given in Table 3, and the additives were added at room temperature.

Visual assessment (Fig. 5) of the bitumen modified with the recycled

plastics during manual mixing showed a high heterogeneity amongst the additives (P1, as mentioned above, and P2 and P3, which did not completely dissolve in the bitumen). Only additives P4 and P5 dissolved completely in the bitumen despite the low mixing energy (additives suitable for the wet process), making them also suitable for addition via the dry process.

Elsewhere, the results of the full experimental testing of the study developed by the authors showed that the P4 and P5 additives produced similar results in bitumen properties, regardless of whether the recycled plastics were added to the bitumen manually or mechanically. Due to the heterogeneity of the resulting bitumens when the P1, P2 and P3 additives were added manually, no further results on the effects of the recycled plastics (added manually) on the bitumen are shown in this study.

In a second step, the addition of recycled plastics into asphalt mixtures was evaluated to further investigate the effects of recycled plastics on the behaviour of asphalt mixtures (fifth phase).

Table 3	
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7 - Ct t	A	- C 1 1			temperature/t	*
Soffening	remperature	of recycled	Diastics and	mixing	remperature/f	ime.

ID	Bitumen	Recycled plastics		Conditions considered	Conditions considered in the study			
	Classification	Main plastics in the	Softening temperature	Mixing temperature recommend by the	Mixing temperature (Mixing time (min.)		
		additive	(°C)	manufacturers (°C)	°C)	Manual	Mechanical	
B1	50/70	_	_	-	155 ²⁾	_	_	
B2-	PMB 45/	-	-	-	165 ³⁾	-	_	
REF	80-65							
B1P1	50/70	LDPE, HDPE, PP	160 - 180	170 – 190	180	1	5	
B2P2	PMB 45/	LDPE, HDPE	>120	173 ¹⁾	173		60	
B2P3	80-65		>100	173 ¹⁾				
B2P4			116	170 – 190			30	
B2P5		PP	156	170 – 190				



Fig. 4. Smooth rod (left) and mixing device for bitumen/recycled plastic (right).



Fig. 5. Visual appearance of the bitumen after manual mixing with recycled plastics.

When adding recycled plastics to asphalt mixtures via the dry process, it should be noted that the addition of P4 and P5 additives, which can be added via the wet process as previously mentioned, can cause rapid melting of the recycled plastics when in contact with the hot aggregates, which can result in these recycled plastics being completely absorbed by the pores of the aggregates and not performing their function. To avoid this, the addition of recycled plastics to asphalt mixtures via the dry process described in Fig. 6 considers two methods, one for additives that can only be added via the dry process (additives P1, P2, P3) and one for additives that can be added via the wet and dry processes (P4, P5):

- For the recycled plastics (P1, P2, and P3), after addition and oneminute mixing with the hot aggregates, the mixture was left in the oven for 30 min to promote softening of the plastics before they were added and mixed with the filler and bitumen for about 5 min.
- For the recycled plastics (P4 and P5), the hot aggregates, filler and bitumen were first mixed for about one minute to ensure complete

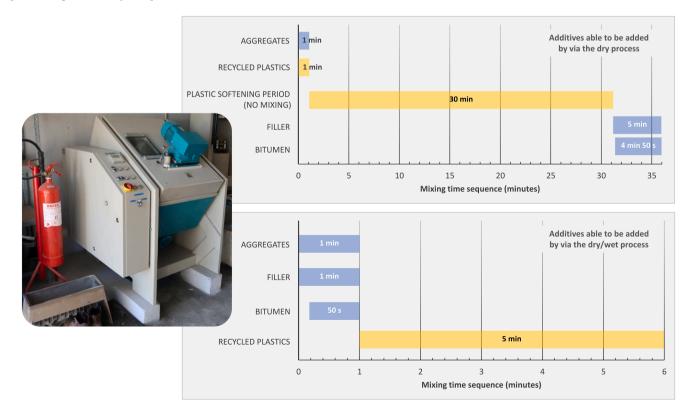


Fig. 6. Mixing device and procedures used in the study to add recycled plastics to asphalt mixtures via the dry process.

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coverage of the materials with the bitumen; then the recycled plastics were added and mixed for 5 min.

The mixing temperatures were the same as in Table 3, and the additives were added at room temperature.

In addition to evaluating the dispersion of the recycled plastics in the asphalt mixtures, recovery of the bitumen was performed for all asphalt mixtures tested using a solvent solution (perchloroethylene) and centrifugation (EN 12697-1). Fig. 7 shows an example of the aggregate of one of the asphalt mixtures studied, which were retained by different sieves (6.3 mm, 2 mm, 1 mm, 0.500 mm, 0.250 mm, 0.125 mm, 0.063 mm and the rest below 0.063 mm, since the size of the recycled plastics is below 5 mm). Fig. 7 is supplemented by a visual example of the cellulose fibers retained in the lower size dimension sieves (≤ 2 mm) after bitumen recovery using the same process as in this study.

According to Fig. 7, no plastic-coated aggregates or individual recycled plastics (for all recycled plastics used in the study) were visible in the aggregates, as is easily the case with cellulose fibers. The process by which the recycled plastics (mainly LDPE, HDPE and PP) were introduced into the asphalt mixture in the laboratory (via the dry process) appears to be suitable.

However, the possible fragmentation of the recycled plastics into micro and nano plastics during production, as well as the low content of recycled plastics in the mix (0.1% to 0.3% of the total mass of the asphalt mixture), could also make the visibility of the plastics difficult. Further studies should be considered to evaluate the influence of the solvent used on the properties of the recycled plastics, as well as the evaluation of the micro and nano plastics generated during the production and application of the asphalt mixture.

2.3. Experimental test programme methodology

An experimental test programme was developed for the fourth and fifth phases (Fig. 3) to investigate the influence of the recycled plastics

on the behaviour of the bitumen, on the affinity between aggregate and bitumen, and on the behaviour of the asphalt mixture (mechanical and surface properties):

- Bitumen was tested for properties such as penetration at a test temperature of 25 °C (EN 1426), softening temperature by the ring and ball test (EN 1427), elastic recovery (EN 13398), cohesion energy (EN 13589) at a test temperature of 5 °C, 10 °C and 15 °C (the latter if the test cannot be completed at 10 °C), ageing resistance by rolling thin film oven test, RTFOT (EN 12607-1) and Brookfield viscosity with temperatures between 100 °C and 220 °C.
- Affinity between aggregate and bitumen according to the procedure described in EN 12697-11 (method A). The test temperature was 5 °C. Visual assessment of the samples was performed after 6 and 24 h. Recycled plastics were added and manually mixed with the hot aggregates for about one minute using a smooth rod as described in Table 3 to simulate reduced interaction between bitumen and plastics.
- The Marshall properties (stability, S; flow, f and quotient, Q) were evaluated according to EN 12697–34 at a constant temperature of 60 °C. The cylindrical specimens were previously conditioned in water at a temperature of 60 °C for 40 min.
- Water sensitivity according to EN 12697–12 (method A) using indirect tensile strength of dry specimens (ITSDRY), indirect tensile strength of wet specimens (ITSWET) and indirect tensile strength ratio (ITSR) of cylindrical test specimens of asphalt mixture (EN 12697–23) at a constant test temperature of 15 °C. The specimens were divided into two groups, one previously conditioned in air (20 °C) and the other previously conditioned with vacuum and water (40 °C), both groups for 72 h.
- Resistance of asphalt mixtures specimens to permanent deformation (EN 12697–22, wheel-tracking device) under repeated passes (10 000 cycles) of a loaded wheel at a constant temperature of 60 °C. The test specimens were conditioned in air during testing (method B).



Fig. 7. Visual aspect of the aggregates for different sieves without the presence of recycle plastics.

• Surface properties in terms of macrotexture depth using a volumetric patch according to EN 13036–1 and slip/skid resistance using the British pendulum (EN 13036–4).

In the laboratory, cylindrical specimens were prepared and compacted according to EN 12697–30 (Marshall compactor, 50 blows per side, diameter 101.6 mm) to evaluate Marshall properties and water sensitivity and according to EN 12697–33 to evaluate resistance to permanent deformation and surface properties (specimen dimensions $300 \text{ mm} \times 260 \text{ mm} \times 50 \text{ mm}$).

3. Influence of recycled plastics on the behaviour of bitumen

Table 4 shows the properties of the bitumen (without/with recycled plastics, mechanical mixing) in terms of penetration, softening temperature, elastic recovery, cohesion energy of the bitumen at 10 $^{\circ}$ C and ageing resistance with RTFOT, which is graphically supplemented by Fig. 8 to Fig. 12.

The results in Table 4 and Fig. 8 show that regardless of the base bitumen used (50/70 or PMB 45/80–65), a decrease in penetration (more pronounced for B1P1 and B2P2) and an increase in softening temperature (more pronounced for B2P2 and B2P4) are observed for all bitumens with additives. In the case of B1P1 added to bitumen 50/70, there was a very significant decrease in penetration as well as an increase in softening temperature, but in the latter case not sufficient to reach the value of the reference bitumen PMB 45/80–65.

A similar elastic recovery (Fig. 9) was observed for all bitumens with additives compared to the reference bitumen PMB 45/80–65, except for B2P2, where a decrease was more pronounced. For bitumen with P1, the elastic recovery was zero, although the base bitumen is a 50/70.

Regarding the cohesion energy (Fig. 9) at a temperature of 10 °C, B2P3 was found to have similar values to the base bitumen (PMB 45/ 80–65) or improved values (B2P4 and B2P5). For B1P1 and B2P2, the test could not be successfully completed, even at 15 °C. It should be noted that the evaluation was not possible to be completed for all bitumens at 5 °C, including the reference bitumen. In addition, the B1P1 showed abnormal behaviour at 5 °C with high stiffness and brittleness, similar to glass. Therefore, the P1 additive is not expected to be suitable for cold climates and/or surface layers. amongst the different bitumens with additives, B2P3 was the only one that generally behaved similarly to the respective base bitumen for the different properties evaluated, and it can be assumed that it acts only as an aggregate and/or as a modifier of the asphalt mixture. Note that the recycled plastics that composed the additive looked more aged and spent then the rest (Fig. 2), which may affect a suitable modification of the bitumen.

In addition, amongst the different bitumen properties studied, a significant linear relationship between penetration and elastic recovery was found (see Fig. 10).

As shown in Fig. 10, an increase in penetration corresponds to an increase in the observed elastic recovery. In general, however, the

addition of recycled plastics should contribute to a potential improvement in the behaviour of the bitumen in terms of permanent deformation resistance without a significant change in elasticity (B2P3, B2P4 and B2P5). It should be noted that in the case of B1P1, the use of the plastic resulted in an improvement in behaviour compared to the base bitumen (50/70), but did not provide the same or improved properties compared to the reference bitumen (PMB 45/80–65).

The progressive ageing of bitumens result of the loss of the volatile fraction of the bitumen (maltene) and/or its transformation into a solid fraction (asphaltene), which usually leads to a decrease in the penetration and increase of the softening temperature and consequently in the stiffness of the bitumen, contributing to an increase in resistance to permanent deformation and to a decrease in fatigue resistance. A trend of this behaviour can be seen in Fig. 12 (complemented by Fig. 11), where a linear relationship is observed between the penetration and the softening temperature of the aged bitumens. Once again, B2P3 was the only one that generally behaved similarly to the respective base bitumen and it can be assumed that it acts only as an aggregate and/or as a modifier of the asphalt mixture.

According to the results, the addition of recycled plastics is not expected to reduce or improve the ageing resistance of the bitumen and thus its resistance to fatigue cracking (especially for B1P1 and B2P2). In terms of resistance to permanent deformation, an improvement in behaviour is predictable, especially for B2P2, similar to what was previously observed due to lower penetration compared to PMB 45/80–65. It would be expected that bitumen B1P1 would also show a predictable improvement in resistance to permanent deformation if it could be evaluated.

In addition, Fig. 13 shows the viscosities of the bitumens with the recycled plastic additives for different temperatures ($100 \degree C$ to $220 \degree C$) compared to the base bitumen (50/70 in the case of P1 and PMB 45/80-65 for the other additives).

Fig. 13 shows that, as expected, recycled plastics cause an increase in bitumen viscosity at the different test temperatures compared to the respective base bitumen. This increase is particularly evident for B1P1, B2P2 and B2P3. Although B2P4 and B2P5 (additives suitable for the wet process) allow a slight increase in viscosity, their viscosity behaves relatively similar to that of the base bitumen (PMB 45/80–65).

4. Influence of recycled plastics on the affinity between aggregate and bitumen

Fig. 14 shows the results of the adhesion loss between the bitumen and the aggregate/recycled plastic additives for 6 and 24 h. The results additionally show the evaluation of the bitumen B1 (50/70) without additives.

The results in Fig. 14 show that the affinity between aggregate and bitumen is improved when B2P2, B2P4 and B2P5 are used compared to the reference bitumen (PMB 45/80–65), and may be considered in further studies to evaluate their potential as adhesion promoters for

Table 4

Result	s of	the	properties	of the	he l	bitumen	without,	/with	recycl	ed	plastics.

Property		Without recycled plastics		With recycled plastics				
		50/70	PMB 45/80-65	50/70	PMB 45/80-65			
		-		LDPE, HDPE, PP	LDPE, HDPE			РР В2Р5
	B1	B2-REF	B1P1	B2P2	B2P3	B2P4		
Penetration, 25 °C (0.1 mm)		60	48	13	26	47	43	37
Softening temperat	ure (°C)	51.6	72.6	64.7	99.6	74.0	89.0	76.9
Elastic recovery, 25	5 °C (%	_1)	88	0	73	87	85	86
Cohesion energy, 1	$0 ^{\circ}\mathrm{C} (\mathrm{J/cm}^2)$	_1)	4.5	_1)	_1)	4.7	5.2	5.3
RTFOT, 163 °C	Penetration, 25 °C (0.1 mm)	_2)	36	_2)	19	34	32	30
	Softening temperature (°C)	_2)	75.6	_2)	101.5	75.4	91.5	87.5
	Mass loss (%)	_2)	0.4	_2)	0.3	0.3	0.3	0.4

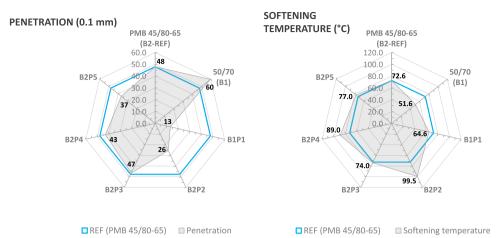


Fig. 8. Influence of the recycled plastics in the penetration and softening temperature of the bitumen.

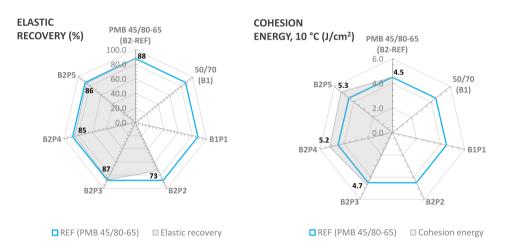


Fig. 9. Influence of the recycled plastics in the elastic recovery and cohesion energy of the bitumen.

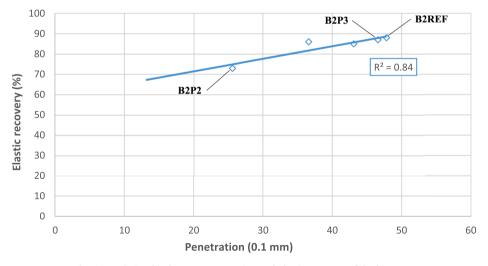


Fig. 10. Relationship between penetration and elastic recovery of the bitumen.

lower quality bitumens.

The unmodified bitumen 50/70 (B1) with and without the addition of recycled plastics (P1) had lower affinity values than the other additives added to the modified bitumen PMB 45/80–65 (B2-REF). In addition, B1P1 decreased the affinity compared to the base bitumen (50/70, B1).

No clear significant relationship was found between the bitumen properties and the affinity between aggregate and bitumen. In addition, the samples with additive P2 occasionally exhibited brittle interfaces between the aggregates and the recycled plastics during testing, as can be seen in Fig. 15, possibly due to the reduced ability to dissolve in the bitumen (see Fig. 5). The results support what was stated above (Section

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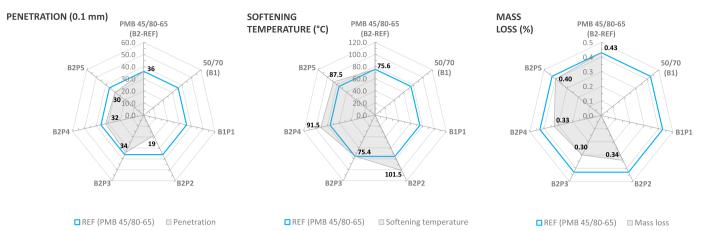


Fig. 11. Influence of recycled plastics on the ageing resistance of the bitumen.

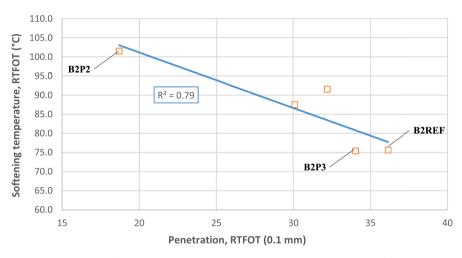


Fig. 12. Relationship between penetration and softening temperature of the aged bitumen.

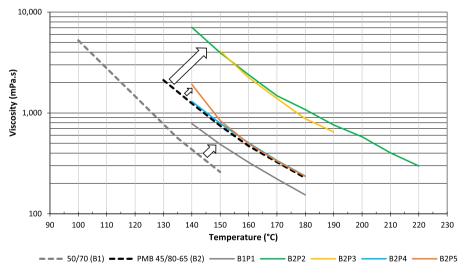
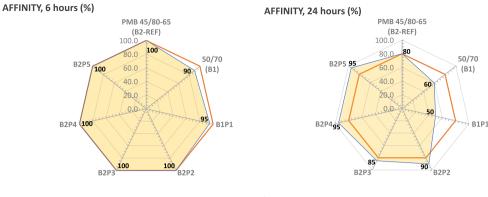


Fig. 13. Influence of the recycled plastics in the viscosity of the bitumen.

2.2). Since the additives P4 and P5 liquefy immediately upon contact with the hot aggregates, it is advisable to add them after the bitumen to prevent their absorption by the porous of the aggregates, which would limit their contribution to improving the performance of the asphalt mixtures.

Regardless of the different degrees of dispersion of the plastics in the bitumen for the different additives, the results show no negative influence on the affinity behaviour, possibly due to the low plastic content (2% to 6% of the total bitumen content).



REF (PMB 45/80-65) Affinity

Fig. 14. Influence of the recycled plastics on the affinity between aggregate and bitumen.

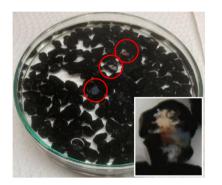


Fig. 15. Aggregate/recycled plastics uncovered with bitumen after 6 h.

5. Influence of recycled plastics on the behaviour of asphalt mixture

The experimental test programme to investigate the influence of the recycled plastics on the behaviour of the asphalt mixture AC 14 surf (mechanical and surface properties) is presented below.

5.1. Marshall properties

Table 5 shows the volumetric properties of the Marshall test specimens, and Fig. 16 shows the results of Marshall properties, such as Marshall stability (S), flow (f), and quotient (Q, ratio between stability and flow) for the asphalt mixtures AC 14 surf.

According to Table 5, the use of recycled plastics in the asphalt mixture tends to increase the porosity of the test specimens. This is possibly due to a reduction in the workability of the asphalt mixture as a result of the lower bitumen penetration and the increase in softening temperature when recycled plastics (see Section 4) are used compared to the reference bitumen (PMB 45/80–65).

Table 5

Volumetric properties of the Marshall test specimens.

Asphalt mixture	ID	Main plastics in the additive	Vv (%)	VMA (%)	VFB (%)
AC 14 surf	B2- REF	-	2.4	14.5	83.2
	B1P1	LDPE, HDPE, PP	4.6	15.8	71.0
	B2P2	LDPE, HDPE	3.9	15.0	74.3
	B2P3		2.9	14.1	79.8
	B2P4		3.5	15.7	77.3
	B2P5	РР	3.0	15.3	80.0

Abbreviations: LDPE - low density polyethylene; HDPE - high density polyethylene; PP - polypropylene; Vv - porosity of a compacted AC 14; VMA - air voids in the aggregate structure; VFB - air voids filled with bitumen.

However, as shown in Fig. 16, the reduction in bitumen penetration due to the use of recycled plastics does not seem to have a significant effect on Marshall stability (between 13.2 kN and 15.4 kN (14.0 kN, reference mixture), which remained similar (B2P4 and B2P5) or increased slightly (B2P2 and B2P3). The only case where the increase can be considered significant is B1P1, where a base bitumen 50/70 was used.

The same trend was observed for the Marshall flow (between 2.6 mm and 3.6 mm (3.3 mm, reference mixture) which tends to remain similar (B2P4 and B2P5) or increase slightly (B2P2 and B2P3), only B1P1 shows a significant decrease in Marshall flow.

Thus, the Marshall quotient generally remains the same, suggesting that the flexibility of the asphalt mixtures, with the exception of B1P1, was not significantly affected by the use of recycled plastics.

Furthermore, no clear significant relationship was found between bitumen properties or aggregate/bitumen affinity and Marshall properties.

5.2. Water sensitivity

Table 6 shows the volumetric properties of the water sensitivity test specimens, and Fig. 17 shows the results of the water sensitivity evaluation (ITS_{DRY} – indirect tensile strength of dry specimens; ITS_{WET} – indirect tensile strength of wet specimens; and ITSR – indirect tensile strength ratio) for the asphalt mixtures AC 14 surf after 72 h of conditioning.

The results (Table 6 and Fig. 17) show that all asphalt mixtures AC 14 surf had similar indirect tensile strength ratio (ITSR) values between 87% and 95% (90%, reference mixture) despite higher porosity of the asphalt mixtures with recycled plastics compared to the reference mixture. Additives P1 and P3 were used for the asphalt mixtures with higher water sensitivity. Additive P5 was the only additive that showed better performance than the reference mix with an ITSR of 95%.

A slight decrease is observed in the indirect tensile strength (dry and wet specimens) according to Fig. 17. This could be related to the higher porosity of the specimens and not directly to the properties of the recycled plastics. Indirectly, however, the increase in porosity is related to the lower workability of the asphalt mixtures when recycled plastics are added.

The rupture of the specimens did not show any significant defects in the coating, and the tested specimens had fewer broken or crushed aggregates. Thus, rupture essentially occurs in the bond between the bituminous mastic and the coarse aggregates. Furthermore, the results do not seem to confirm the possible negative influence of brittle interfaces (additive P2) on water sensitivity.

In addition, no clear significant relationship was found between bitumen properties or aggregate/bitumen affinity, Marshall properties and water sensitivity properties.



Fig. 16. Influence of recycled plastics on Marshall properties.

Table 6

Volumetric properties of the water sensitivity test specimens.

Asphalt mixture	ID	Main plastics in the additive	Vv (%)	VMA (%)	VFB (%)
AC 14 surf	B2- REF	-	2.5	14.6	82.5
	B1P1	LDPE, HDPE, PP	4.5	15.7	71.4
	B2P2	LDPE, HDPE	3.8	14.9	74.8
	B2P3		2.9	14.2	79.2
	B2P4		2.9	15.1	80.9
	B2P5	PP	3.2	15.4	79.3

Abbreviations: LDPE - low density polyethylene; HDPE - high density polyethylene; PP - polypropylene; Vv - porosity of a compacted AC 14; VMA - air voids in the aggregate structure; VFB - air voids filled with bitumen.

5.3. Resistance of asphalt mixtures to permanent deformation

Table 7 shows the volumetric properties of the permanent deformation test specimens, and Fig. 18 shows the results of the evaluation of resistance to permanent deformation (RD_{AIR} – maximum rut depth, in air, and WTS_{AIR} – maximum wheel tracking slope, in air) for the asphalt mixtures AC 14 surf.

From the results presented in Table 7 and Fig. 18, it can be seen that

all asphalt mixtures AC 14 surf have good resistance to permanent deformation with RD_{AIR} between 1.1 mm and 1.9 mm (1.9 mm, reference mixture) and WTS_{AIR} between 0.02 mm and 0.05 mm (0.04 mm, reference mixture). Despite the good results of the reference mixture, the use of recycled plastics further improved the resistance compared to the reference mixture, even for the asphalt mixture with unmodified bitumen (B1P1), except for the additive B2P5 (which is polypropylene, PP recycled plastics). Although all mixtures have a WTS_{AIR} value far below the specified value (WTS_{AIR} \leq 0.10) for surface layers.

Table 7

Volumetric properties of the permanent deformation test specimens.

-	-	-		-	
Asphalt mixture	ID	Main plastics in the additive	Vv (%)	VMA (%)	VFB (%)
AC 14 surf	B2- REF	-	4.4	16.2	72.9
	B1P1	LDPE, HDPE, PP	5.3	16.4	67.8
	B2P2	LDPE, HDPE	5.9	16.8	64.9
	B2P3		4.5	15.6	70.9
	B2P4		3.4	15.6	77.9
	B2P5	PP	6.1	17.9	66.2

Abbreviations: LDPE - low density polyethylene; HDPE - high density polyethylene; PP - polypropylene; Vv - porosity of a compacted AC 14; VMA - air voids in the aggregate structure; VFB - air voids filled with bitumen.

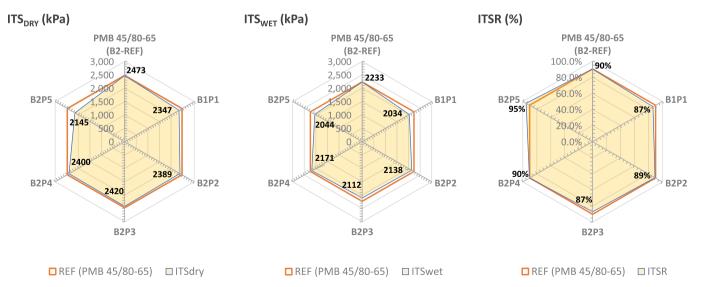


Fig. 17. Influence of recycled plastics on the water sensitivity of asphalt mixtures.

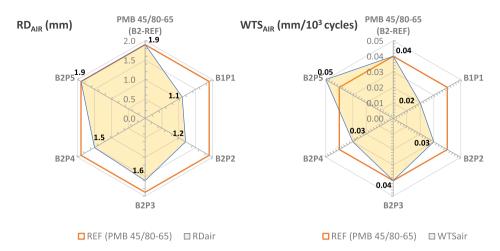


Fig. 18. Influence of recycled plastics on the resistance to permanent deformation of asphalt mixtures.

5.4. Surface properties

Fig. 19 shows the results of the surface properties (macrotexture depth with a volumetric patch and slip/skid resistance with the British pendulum). The test specimens are the same as those previously used to evaluate the resistance to permanent deformation (see Table 7 for volumetric properties of the test specimens).

The results obtained (Fig. 19) show that all asphalt mixtures AC 14 surf had a similar macrotexture between 1.3 mm and 1.6 mm (1.5 mm, reference mixture, B2-REF), similar to a stone mastic asphalt due to gap graded gradation of the mixtures evaluated.

All asphalt mixtures AC 14 surf with recycled plastics had higher slip/skid resistance (between 72 and 83) than the reference mixture [value of 69, above the specified limit in Portugal \geq 60, [10]]. The observed higher slip/skid resistance is not linearly related to the higher porosity or macrotexture of the asphalt mixtures with recycled plastics. Therefore, it could be related to a harder bitumen film thickness around the aggregates on the surface of the asphalt mixture. This conclusion is confirmed by other studies developed by the authors, where the slip/skid resistance of the asphalt mixtures with recycled plastics remained almost the same after removing the bitumen film coating around the aggregates on the surface of the asphalt mixtures.

6. Conclusions

This study investigated the influence of five patented recycled plastic

additives on the behaviour of bitumen and asphalt mixtures and on the affinity between the aggregates and the bitumen.

As a contribution to a better understanding of the use of recycled plastics in pavement construction/rehabilitation, the following main conclusions can be drawn from the results of this study:

Addition of recycled plastics into bitumen/asphalt mixture

- Visual assessment of the bitumen modified with the recycled plastics (manual mixing) showed a high heterogeneity, amongst the additives only capable of being added via the dry process.
- The addition of recycled plastics to asphalt mixtures via the dry process, which can be added via the wet process, can cause rapid melting of the recycled plastics when in contact with the hot aggregates, which may cause these recycled plastics to be completely absorbed by the pores of the aggregates and not performing their function.
- No plastic-coated aggregates or individual recycled plastics (for all recycled plastics used in the study) were visible in the aggregates. The process by which the recycled plastics (mainly LDPE, HDPE and PP) were introduced into the asphalt mixture in the laboratory (via the dry process) may be recommended as suitable.
- Possible fragmentation of the recycled plastics into micro and nano plastics during production, as well as the low content of recycled plastics in the mix, can make the visibility of the plastics difficult in the aggregates. Further studies should be considered to evaluate the influence of the solvent used on the properties of the recycled

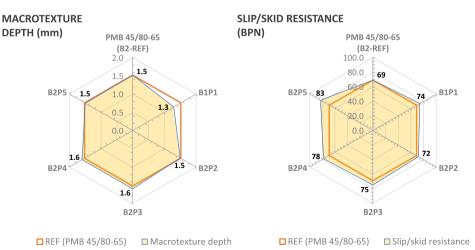


Fig. 19. Influence of recycled plastics on the surface properties of asphalt mixtures.

plastics, as well as the evaluation of the micro and nano plastics generated during the production and application of the asphalt mixture.

Influence of recycled plastics on the behaviour of bitumen (compared to polymer modified reference bitumen)

- Regardless of the base bitumen used, all bitumens with recycled plastics showed a decrease in penetration and an increase in softening temperature. The additive consisting of LDPE, HDPE and PP showed a high ability to modify an unmodified bitumen, but without equivalent properties to the polymer modified bitumen.
- All of the bitumens with recycled plastics had similar elastic recovery, except for one of the additives with LDPE and HDPE, which had a significant decrease in recovery. The additive consisting of LDPE, HDPE and PP could not improve the elastic recovery of an unmodified bitumen.
- A significant linear relationship was observed between penetration and elastic recovery, with an increase in penetration accompanied by an increase in observed elastic recovery.
- Not all bitumens with recycled plastics exhibited similar or improved cohesion energy (the latter applies to both additives that can be added by via the wet process). The additive of LDPE, HDPE and PP was unable to improve the cohesion energy of an unmodified bitumen and showed abnormal behaviour at low temperatures with high stiffness and brittleness, similar to glass, making it potentially unsuitable for cold climates.
- The use of recycled plastics in the form of chips, which may be overaged and spent, may interfere with suitable modification of bitumen properties, as shown by one of the additives used (LDPE and HDPE).
- The addition of recycled plastics should contribute to a potential improvement in the behaviour of the bitumen in terms of permanent deformation resistance, without significantly altering elasticity (additives consisting mainly of LDPE and HDPE or PP).
- The addition of recycled plastics did not improve the ageing resistance of the bitumen and thus its potential fatigue cracking resistance.
- The addition of recycled plastics contributed to an increase in bitumen viscosity, with the exception of additives that can be added by via the wet process, which behave similarly to the base bitumen.
- The addition of recycled plastics leads to an increase in the temperature required to produce asphalt mixtures. The temperature increase may lead to higher energy consumption and exhaust gas emission during asphalt mixture production and application.

Influence of recycled plastics on the affinity between aggregate and bitumen (compared to polymer modified reference bitumen)

- The samples evaluated with recycled plastics show similar or improved behaviour in terms of affinity between the aggregate and the bitumen. The additive consisting of LDPE, HDPE and PP could not improve the affinity of an unmodified bitumen.
- The use of low energy to mix the recycled plastics (for addition via dry process) may occasionally result in brittle interfaces between the aggregate and the plastic. However, no correlation with a decrease in affinity was observed.

Influence of recycled plastics on the behaviour of asphalt mixture (compared to asphalt mixture with polymer modified reference bitumen):

• The use of recycled plastics in the asphalt mixture tends to increase the porosity of the test specimens, possibly due to a reduction in the workability of the asphalt mixture as a result of the reduction in bitumen penetration and the increase in softening temperature.

- Marshall stability and flow generally remain similar (additives for wet process addition) or increased slightly (additives with LDPE and HDPE). The only case where the Marshall stability and flow decreased was the asphalt mixture with the additive LDPE, HDPE and PP with an unmodified bitumen.
- The Marshall quotient generally remained the same, suggesting that the flexibility of the asphalt mixtures was not significantly affected using recycled plastics, except for asphalt mixture with the addition of LDPE, HDPE and PP.
- The water sensitivity (ITSR) of the asphalt mixtures was not significantly affected by the use of recycled plastics (LDPE, HDPE with/without PP), or slightly improved (additive with polypropylene, PP).
- Asphalt mixtures with recycled plastics exhibited similar (or slightly lower, polypropylene only, PP) or improved resistance to permanent deformation (LDPE, HDPE with/without PP).
- Surface properties remained similar for the macrotexture, and the higher slip/skid resistance could be related to a harder bitumen film thickness around the aggregates in the asphalt mixtures with recycled plastics.
- No significant relationship was found between bitumen properties, aggregate-bitumen affinity, Marshall properties, water sensitivity, resistance to permanent deformation, and surface properties.

The addition of recycled plastics into asphalt mixtures via the dry process, regardless of the type, form, plastic content, replacement or increment relative to the bitumen content, as used in this study, shows itself to be an alternative to other techniques for pavement construction and rehabilitation. However, further studies need to be conducted to ensure improved performance in terms of fatigue and ageing resistance of asphalt mixtures, as well as environmental and economic benefits.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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