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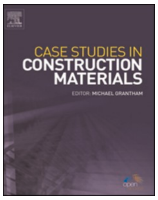

Sustainable asphalt mixtures by partial replacement of fine aggregates with low-grade magnesium carbonate by-product

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ABSTRACT

The utilisation of large quantities of raw materials for asphalt mixtures manufacturing, such as aggregates, is an environmental issue that must be addressed. This concern has led to valorising waste and by-products as more sustainable alternative raw materials. This research is aimed at evaluating the use of a low-grade magnesium carbonate (LG-MC) by-product as a partial replacement of fine aggregates and as a filler of asphalt mixtures. A mechanical analysis has been performed studying the effect of this by-product on the moisture sensitivity, cracking resistance and cohesion loss resistance of asphalt mixtures. Cracking resistance was assessed under different temperatures (20, 5 and −5 °C) and conditions (unconditioned and aged). Results indicated that moisture sensitivity and cohesion loss resistance of asphalt mixtures with LG-MC by-product were not affected, obtaining similar results to those of the reference mixture. A protective effect in the mixture cracking resistance was observed using LG-MC. At low temperatures or after ageing, this by-product tends to maintain ductility to a greater extent. The study indicates that LG-MC is suitable as a partial substitute for the fine fraction of aggregates, as well as for the total amount of filler in asphalt mixtures manufacturing for road pavements.

1. Introduction

Most of the European road and highway network pavements (close to 90%) are made with asphalt mixtures [1]. In 2020, the total production of hot and warm mix asphalt in all the European countries was around 277 million tonnes approximately [2]. Around 90% of asphalt mixtures are composed of natural aggregates of various sizes [3]. The extraction and subsequent consumption of aggregates from quarries have a significant environmental impact, which, together with the fact that natural resources are limited, makes it imperative to reduce their use in order to achieve a lower environmental impact and avoid the depletion of non-renewable resources. A solution to this problem would be to valorise the waste or by-products that end up in landfills for their use in bituminous mixtures manufacturing. This would reduce the consumption of raw materials, and therefore, their total environmental impact [4]. In this regard, several studies to incorporate waste and by-products in pavements have been carried out [5–8].

Some waste or by-products such as reclaimed asphalt pavement (RAP), copper slag, waste glass, steel slag or stabilized bottom ash from municipal solid waste incineration have been studied for their use as a replacement for aggregates in the manufacture of bituminous mixtures, being RAP one of the most used wastes [9–13]. All these studies demonstrated the feasibility of using industrial residues or by-products as more environmentally sustainable alternatives to natural aggregates for bituminous mixtures

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manufacturing.

Other residues or by-products such as rice husk ash, date seed ash, waste glass, waste marble, waste granite, steel slag or hydrated lime powder have been used as filler for asphalt mixtures (the part of the aggregate whose particles pass the sieves between 0.080 and 0.063 mm) [14–16]. Asphalt mixtures made with rice husk ash or date seed ash as replacements for mineral filler increased Marshall stability around 20% and 30%, respectively, as well as the stiffness modulus. Likewise, an improvement in thermal sensitivity and adhesiveness between the aggregate and the binder of the mixtures was observed [14]. Awed et al. (2020) studied the use of marble, granite, steel slag and hydrated lime powder by-products as fillers in asphalt mixtures. The authors found that the mixture with marble showed the greatest stability increasing by 21.6% concerning conventional filler (limestone). The mixture with hydrated lime powder improved the resistance of the mixture against moisture damage, and its behaviour as to dynamic modulus and flow number. On the other hand, the findings of Gedik (2021) indicated that the mixtures with recycled waste glass as filler improved their thermal susceptibility and their behaviour to fatigue for certain applications. An increase of 35% in their resistance to fatigue was found in comparison with the reference mixture.

On the other hand, magnesia (MgO) is a magnesium compound widely used in different productive sectors, both in the primary and secondary sectors. Dead-burned magnesia is the primary component in refractory materials, which are of great industrial value as furnaces and kilns linings, with the steel industry being the largest user of refractory magnesia. Caustic-calcined magnesia is an essential component in several agricultural, environmental, construction and industrial applications. Natural magnesite is the most common source of magnesia through a high-temperature calcination process. The US Geological Survey [17] estimates that the production of magnesite in 2020 reached 26 million metric tons worldwide. Thus, reserves stand, with reserves sitting at 7.6 billion metric tons, with the Spanish company Magnesitas Navarras, S.A. being one of the main European magnesia producers. During the process of beneficiation of natural magnesite and, subsequently, the calcination process to obtain magnesia, different mineral by-products with high magnesium contents are produced [18]. Among them, a by-product with a high potential to be used in the manufacture of bituminous mixtures is the low-grade magnesium carbonate (LG-MC) obtained during the magnesite gravity concentration stage. For decades, this by-product has been stored in the open in heaps near magnesia production plants. This situation currently makes it possible to recover large volumes of this by-product and to properly manage the accumulated volume.

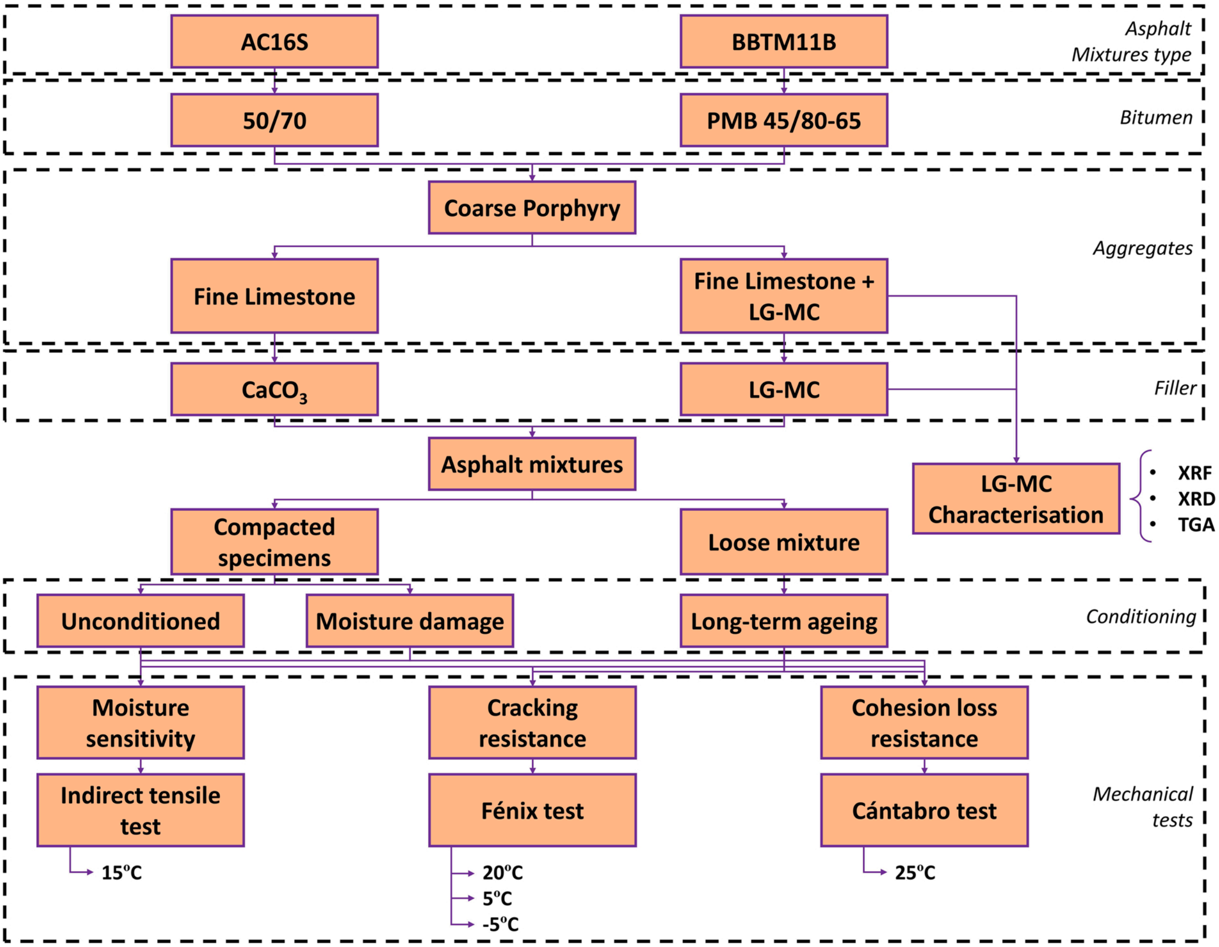

Fig. 1. Flow chart of the experimental study.

López-Montero et al. [19] evaluated the potential use of LG-MC by-product as filler in the manufacture of asphalt mixtures using the UCL method (*Universal de Caracterización de Ligantes*) [20,21]. The results of this analysis showed some positive effects of the LG-MC filler against resistance to ageing, reducing losses to abrasion. However, although it is known that LG-MC has a high potential to be used in bituminous mixtures, it is necessary to validate the effect of the use of this material on the behaviour of bituminous mixtures.

The behaviour of asphalt pavements is complex depending on factors such as temperature, climate, or their proper construction, among others. All of them could cause premature deterioration of the pavement [22–24]. Asphalt mixtures can change their behaviour from ductile to brittle due to the effect of their temperature or their ageing condition [25,26]. For all these reasons, it is crucial to study the mechanical properties of asphalt mixtures both when they are aged and at low temperatures, since mixtures are most susceptible to cracking [27].

This research aims to validate the use of LG-MC as a fine aggregate and filler in asphalt mixtures for road pavements. This has been achieved through a mechanical analysis by replacing conventional calcium carbonate filler and part of the fine aggregates for LG-MC in two different asphalt mixtures manufacturing. To assess how this by-product can affect the behaviour of asphalt mixtures, the moisture sensitivity, the cracking resistance at different temperatures and under different asphalt mixtures conditions (unconditioned and unaged) and the cohesion loss resistance were carried through the indirect tensile strength, the Fénix and the Cántabro tests, respectively.

2. Materials and methods

Fig. 1 shows the flowchart of the research carried out. Two types of bituminous mixtures were studied: a gap-graded one (BBTM11B) (EN 13108–2:2006) and an asphalt concrete semi-dense one (AC16S) (EN 13108–1:2016). The first one was made with a polymer-modified binder, PMB 45/80–65, while the semi-dense mixture was made with a conventional binder, 50/70. In both mixtures, the coarse aggregate was porphyry. The fine aggregate was partially replaced by LG-MC, using this by-product also as a filler for their manufacture. Results were compared with those obtained for the same type of mixture entirely made of coarse porphyry aggregate, fine limestone aggregate and calcium carbonate filler. Asphalt mixtures were subjected to both ageing and moisture damage to evaluate their behaviour against these conditions. Unconditioned and subjected to moisture damage specimens were tested through the indirect tensile strength test to evaluate their water sensitivity. Unconditioned and long-term aged specimens were tested through Fénix at the temperatures of 20, 5 and − 5 °C to evaluate the cracking resistance of the mixture. Finally, the unconditioned, subjected to moisture damage and long-term aged specimens were tested under Cántabro to evaluate the mixture cohesion loss resistance. At least four replications were performed for each study condition.

2.1. Materials

2.1.1. Aggregates

Coarse porphyry and fine limestone aggregates were used for asphalt mixtures manufacturing. Fine aggregates were partially replaced by LG-MC while filler was entirely LG-MC. Commercial calcium carbonate was used as reference filler. According to the calcium carbonate chemical composition, the 98.96% of it is $CaCO_3$.

2.1.2. Raw material characterisation

Fine aggregates and filler for the asphalt mixture manufacturing were partially replaced by a low-grade magnesium carbonate (LG-MC) by-product. This by-product was supplied by the company Magnesitas Navarras, S.A., located in Zubiri (Navarra, Spain), generated during the beneficiation carried out by the sink-float separation process using ferrosilicon aqueous suspension as the dense media. Approximately 80,000 tonnes of this by-product are currently generated each year, which are stored in a heap. Thus, it is estimated that more than one million tonnes of this by-product are accumulated after 75 years of activity. Table 1 shows the particle size distribution of the LG-MC obtained according to the UNE-EN 933–1 standard.

A Panalytical Philips PW2400 sequential spectrometer with UniQuant V5.0 software was used to obtain the X-ray fluorescence (XRF) spectroscopy of LG-MC. Furthermore, its crystalline phases were determined from X-ray diffraction (XRD), using a PANalytical X'Pert PRO MPD alpha1 powder diffractometer with CuKα1 radiation ($\lambda = 1.541$ A), work power of 45 kV-40 mA, 4–100° 2θ range, 0.0263° step size and 100 s measuring time. Finally, the behaviour of the by-product at high temperatures was analysed by a thermogravimetric analysis (TGA) using a TA Instruments SDT Q600 device in a nitrogen atmosphere (50 ml·min^{-1}). The heating rate was 10 °C·min^{-1} up to 1200 °C.

2.1.3. Asphalt binders

A conventional bitumen, 50/70, and an SBS polymer-modified one, PMB 45/80–65, were used for the manufacture of asphalt mixtures. Table 2 shows the main properties of both bitumens.

Table 1
Particle size distribution of the LG-MC by-product according to the UNE-EN 933–1 standard.

Sieve (mm)	4	2	0.5	0.25	0.125	0.063
Passing (%)	100.0	98.9	98.2	97.6	84.6	60.5

2.2. Asphalt mixtures

Two different types of bituminous mixtures were used: a gap-graded mixture (BBTM11B) and a semi-dense concrete asphalt mixture (AC16S). Considering the particle size distribution of LG-MC and the aggregates particle size distribution of the selected asphalt mixtures (Fig. 2), the total percentage of LG-MC was 12.3% in the case of the BBTM11B mixture and 11.6% for the AC16S mixture. This percentage includes the total amount of filler for asphalt manufacturing and a part of the finest fraction of aggregates. The filler content was the optimum filler/bitumen ratio presented in the characterisation study of LG-MC as a filler (López-Montero et al., 2022). This ratio was obtained by a volumetric dosage procedure from the relationship between the volumetric concentration and the critical concentration of the filler. The optimal filler/bitumen ratio was 1.33 for the calcium carbonate filler and 1.49 for the LG-MC filler.

The binder content was 4.75 wt% of mixture for the BBTM11B, while in the case of the AC16S asphalt this content was 4.5 wt% of mixture. Cylindrical specimens with 101.6 mm of diameter and 63.5 mm height, approximately, were prepared (Marshall specimens). They were compacted by impact. For the BBTM11B mixture, 50 blows per side were applied for its compaction. For the AC16S mixture, specimens for the Fénix test were compacted by applying 75 blows per side while specimens for the water sensitivity analysis by the indirect tensile strength test were compacted by applying 50 blows per side. The bulk density and air voids content of asphalt mixture specimens were determined according to standards EN 12697–6 and EN 12697–8, respectively.

2.3. Asphalt mixtures conditioning

The water sensitivity study of the specimens from the indirect tensile strength test was carried out following the EN 12697–12 specification. For this, the group of wet specimens was placed in a water bath at 40 ºC for a period of 72 h. Previously, the specimens were subjected to a vacuum of 6.7 kPa for 30 min. Both types of asphalt mixtures (gap graded, BBTM11B, and semi-dense concrete, AC16S, were subjected to this conditioning).

For the Cántabro test, the moisture damage procedure (NLT-362/92) consisted of placing the Marshall specimens of gap-graded asphalt mixture (BBTM11B) in a water bath at 60 ºC for 24 h. Afterwards, the specimens were removed from the bath and conditioned at a temperature of 25 ºC to test them after 24 h.

The specimens were aged according to the long-term ageing procedure established by the RILEM committee [28]. This consists of ageing the specimens in the short term, leaving the mixture loose in an oven at 135 ºC for 4 h. Once the specimens are short-term aged, they are left in the oven at 85 ºC for 9 days. The mixture is stirred three times with a time interval of at least 1 day between stirrings. Once aged, the mixture specimens are compacted.

2.4. Mechanical tests

The analysis of the behaviour of mixtures partially manufactured with LG-MC was done through the study of sensitivity to water, resistance to cracking and resistance to abrasion losses. To do that, the indirect tensile strength, Fénix and Cántabro tests were used, respectively. The Cántabro test was carried out only on specimens of the BBTM11B asphalt mixture.

2.4.1. Indirect tensile strength test

To determine the water sensitivity of asphalt mixtures, the indirect tensile strength (ITS) test according to EN 12697–23 was used. Marshall specimens were subjected to a diametral load in the direction of the specimen axis at a constant displacement speed of 50 ± 2 mm/min until breaking. Two groups of specimens, unconditioned and after moisture conditioning (wet), were tested at a temperature of 15 ºC. From this test, ITS of asphalt mixtures specimens was determined, Eq. 1:

$$ITS = 2 \cdot P / (\pi \cdot D \cdot H) \tag{1}$$

where *ITS* is the indirect tensile strength (GPa), *P* is the maximum load (kN), *D* is the specimen diameter (mm), and *H* is the height of the specimen (mm).

Table 2
Characteristics of bitumens (50/70 and PMB 45/80–65) both the original bitumen and the residue after ageing.

Characteristic	Standard	Bitumen 50/70	Bitumen PMB 45/80–65
Original Bitumen			
Penetration at 20°C (0.1 mm)	EN 1426	61	57
Softening point R&B (°C)	EN 1427	50.9	65.3
Fraass breaking point (°C)	EN 12593	-14	-15
Flash point (°C)	EN 2592	280	290
Residue after ageing			
Mass variation (%)	EN 12607–1	0.10	0.29
Penetration at 25°C (% p.o.)	EN 1426	66	64
Δ Softening point (°C)	EN 1427	7.6	10.0

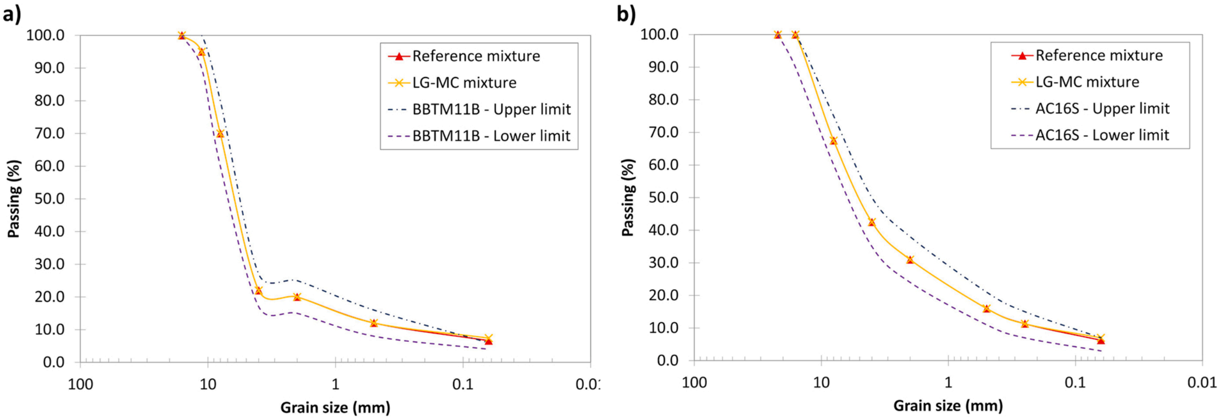

Fig. 2. Aggregate particle size distribution of the reference and the asphalt mixture partially manufactured with LG-MC a) BBTM11B and b) AC16S.

2.4.2. Fénix test

Fénix test (NLT-383/20) is a direct tensile monotonic test at a constant displacement speed (1 mm/min). It consists of applying a deflected tensile force to a semi-cylindrical specimen by means of two steel plates glued on the specimen diametral plane. The steel plates are attached to the press by ball joints (Fig. 3). This test can be performed at different temperatures. Specifically, in this research, the Fénix test was carried out at the temperatures of 20, 5 and − 5 °C, and for the mixtures unconditioned and long-term aged (LTA).

The applied load is recorded during the test depending on the displacement (Fig. 3). From the load-displacement curve, it is possible to obtain various parameters related to the mechanical and resistant characteristics of the mixtures. However, to determine the behaviour of the mixture with LG-MC compared to the reference one, this study focused on the test parameters most relevant for this purpose. These are the maximum strength (RT), the tensile stiffness index (IRT) and the toughness displacement (DT).

The maximum strength, RT, is related to the asphalt mastic cohesion and is determined by Eq. 2.

$$RT = F_{\max}/S \tag{2}$$

where RT is the maximum strength (MPa), F_{max} is the peak load (kN), and S is the cross-sectional area (mm^2). This area is obtained as the product of the height of the specimen by the reduced radius (radius measured in the induced crack zone subtracting its depth).

The tensile stiffness index, IRT, is obtained as the slope of the load-displacement curve between 25% and 50% of the peak load in the loading phase divided by the cross-sectional area. This index is related to the modulus of the mixture and is determined by Eq. 3.

$$IRT = 1000(F_{50} - F_{25})/(S \cdot (d_{50} - d_{25})) \tag{3}$$

where IRT is the tensile stiffness index (MPa/mm), F_{50} and F_{25} are the values corresponding to half and a quarter of the peak load in the loading phase (kN), respectively, S is the cross-sectional area (mm^2) and, d_{50} and d_{25} are the displacement at 50% and 25% of the peak load in the loading phase (mm), respectively.

Finally, the ductility of the mixture could be evaluated by the toughness displacement (DT) which is determined by subtracting the displacement at the peak load (mm), d_{max}, to the displacement corresponding to 50% of the peak load in the cracking phase (mm), $d_{50\,pm}$, Eq. 4.

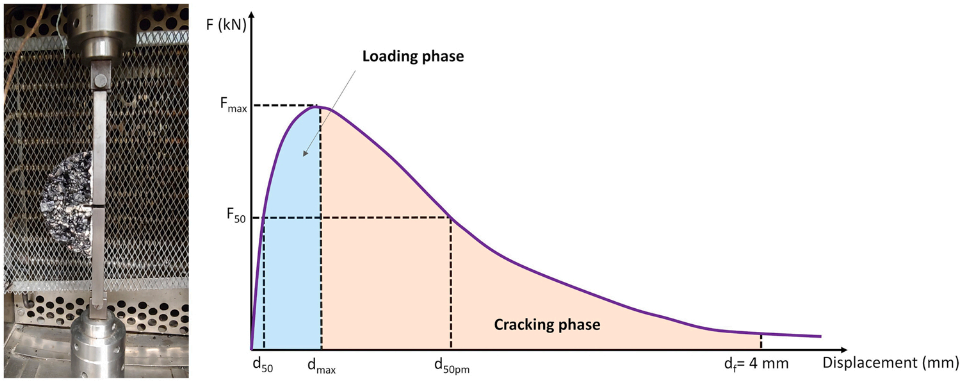

Fig. 3. On the left, Fénix test setup on the testing machine. Steel plates where the semi-cylindrical specimen is glued are attached to the press. On the right, load-displacement curve obtained from the Fénix test (adapted from NLT-383/20).

$$DT = d_{50pm} - d_{\max} \tag{4}$$

2.4.3. Cántabro test

The Cántabro test (NLT-352/00; EN 12697–17) was carried out on the BBTM11B mixture. This test makes it possible to determine the resistance of bituminous mixtures with open grading to the traffic shredding forces. It consists of determining the weight loss (wt%) suffered by a Marshall specimen after being subjected to 300 revolutions in the Los Angeles abrasion machine, without any type of abrasive load. Losses were obtained for the mixture unconditioned, subjected to moisture damage and long-term aged (LTA). Cántabro test was carried out at 25 °C.

3. Results and discussion

3.1. Low-grade magnesium carbonate characterisation

The elemental composition of LG-MC determined by XRF is given in Table 3. Major elements are expressed as their most stable oxide. As expected, magnesium is the main element, followed by calcium. The higher value of the loss of ignition (LOI) parameter, carried out at 1100 °C, was due to the presence carbonates that decompose below 1000 °C.

Fig. 4 shows the X-ray diffraction (XRD) pattern of LG-MC. The main crystalline phases identified were magnesite ($MgCO_3$; PDF# 01–078–2442) and dolomite ($CaMg(CO_3)_2$. Other phases such as quartz (SiO_2; PDF# 01–078–2315) and calcite ($CaCO_3$; PDF# 01–070–0095) were identified as minor phases. Understandably by the nature of LG-MC, these results were consistent with the obtained in Table 3. Due to the presence of quartz, which has a lower specific surface area than magnesite and dolomite, and an acidic character, the adhesion between LG-MC and binder could be affected. This fact may influence the performance of asphalt mixtures against moisture damage.

The thermal decomposition of LG-MC is observed in Fig. 5. Two stages are observed in the graph: the first between the temperature range of 539ºC and 682ºC, and the second between the range of 682ºC and 777ºC. According to the XRD results, the first would correspond to the decomposition of magnesite, while the second one corresponds to the decomposition of dolomite.

3.2. Asphalt mixtures testing

3.2.1. Indirect tensile strength test

The indirect tensile strength test after immersion evaluates the water sensibility of the mixtures, which is a general requirement for bituminous mixtures included in the standard EN 13108. From the results, the aggregate-bitumen adhesiveness is evaluated, analysing the effect of the action of water on the degradation of the mixture. Table 4 shows the bulk density and air voids content of the specimens under the different conditioning (unconditioned and wet), tested by indirect tensile strength. The order of magnitude of air voids content for the mixtures made with the reference filler and the mixtures with LG-MC is similar. This makes the results comparable.

Fig. 6 shows the results of the indirect tensile strength (ITS) for the gap-graded mixture, BBTM11B, as well as for the semi-dense mixture, AC16S, in both conditions: unconditioned and wet, respectively. It is observed that ITS values are slightly lower in the mixtures with LG-MC. However, taking into account test dispersions, it can be considered that the differences between the mixture with the reference filler and the mixture with LG-MC are not significant. Table 5 shows the results of the ANOVA analysis. They verify that no significant differences are observed between the ITS results of the mixture with the reference filler and the mixture with LG-MC for each of the conditions studied. Likewise, when obtaining the indirect tensile strength ratio values (the ratio of the ITS wet specimens versus the ITS unconditioned specimens), it is observed that they are similar between the mixtures with the reference filler and the mixtures with LG-MC (Fig. 7).

For all the cases studied, it is observed that water does not negatively affect the ITS values, obtaining values of indirect tensile strength ratio above 100%. The increase in ITS values after moisture damage could be due to the way of conditioning the mixture. The water bath temperature to subject the mixture to moisture damage may be causing small ageing of the binder, which would slightly increase its stiffness and, therefore, the mixture's indirect tensile strength after water conditioning [29].

Although quartz, which could affect the aggregate-bitumen adhesion, was detected as a crystalline phase of LG-MC, it is demonstrated that the resistance of the mixtures against moisture damage is not influenced by this fact. So, magnesite and dolomite rule the performance of the mixture. Thus, the mixtures made with LG-MC as filler and part of fine aggregates showed a similar behaviour than the reference one, normally used in wearing courses of pavements.

Table 3
Elemental composition of LG-MC from an X-ray fluorescence.

Oxides	MgO	CaO	SiO_2	Fe_2O_3	Al_2O_3	MnO	K_2O	P_2O_5	TiO_2	Na_2O	LOI
wt. (%)	34.4	11.5	4.92	1.62	0.41	0.09	0.08	0.07	0.02	0.02	46.9
SD	0.090	0.106	0.038	0.022	0.001	0.001	0.001	0.000	0.001	0.001	0.257

LOI: Loss of ignition at 1100 °C.

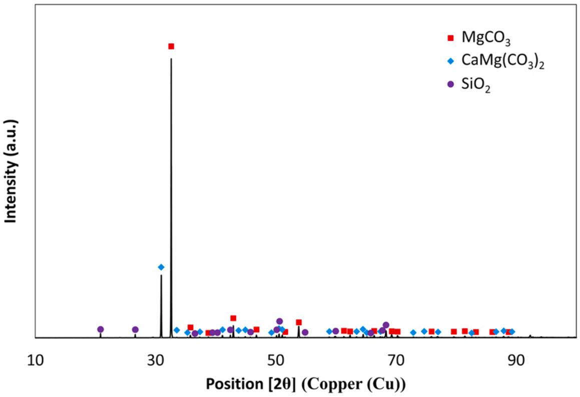

Fig. 4. LG-MC X-ray diffraction results to determine its crystalline structure. Magnesite ($MgCO_3$), dolomite ($CaMg(CO_3)_2$) and quartz (SiO_2) are highlighted.

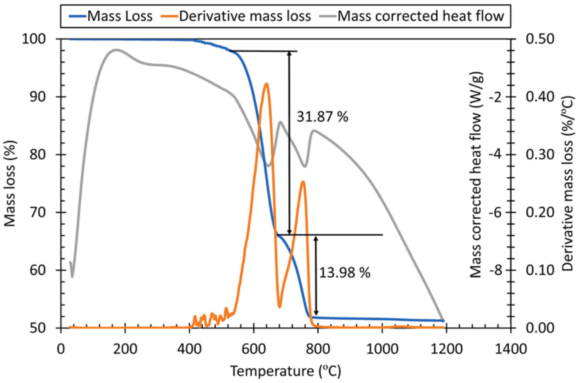

Fig. 5. LG-MC TGA thermal decomposition. Mass loss (y-axis), mass corrected heat flow (secondary y-axis, on the left) and derivative mass loss (secondary y-axis, on the right).

Table 4
Bulk density and voids content average values of the specimens used for the indirect tensile strength test.

Mixture		Condition	Bulk density (g/cm^3)	Voids content (%)
BBTM11B	Reference	Uncond	2.122	15.3
		Wet	2.116	15.5
	LG-MC	Uncond	2.120	16.1
		Wet	2.134	15.5
AC16S	Reference	Uncond	2.406	4.4
		Wet	2.399	4.7
	LG-MC	Uncond	2.391	5.1
		Wet	2.391	5.1

3.2.2. *Fénix test*

Table 6 shows the average values of bulk density and air voids content of the specimens tested at Fénix for the two types of mixtures (BBTM11B and AC16S), the different conditions considered (unconditioned and long-term aged) and for the three test temperatures (20, 5 and −5 °C). Just as the specimens subjected to the indirect tensile strength test, the order of magnitude in all cases studied is the same which make the results of the test comparable. The greatest differences are observed in the AC16S mixture, especially after submitting them to ageing. These differences are expected due to the major difficulty of compacting this mixture after ageing. In this case, the higher voids content observed in the mixture with LG-MC would make it more critical in terms of the properties evaluated.

Fig. 8 shows the results of the maximum strength (RT) for the two types of asphalt mixture (BBTM11B and AC16S), unconditioned and after long-term ageing (LTA), for the test temperatures of 20, 5 and − 5 °C. For both types of mixture, the ageing tends to increase RT at the different test temperatures regardless of the filler type. Likewise, it is observed that at medium temperatures (20 °C), the behaviour of each of the mixtures considered is similar, regardless of whether LG-MC has been used for their manufacturing. At lower temperatures (5 and −5 °C), irrespective of their ageing condition, the RT of the BBTM11B mixture tends to be higher when LG-MC is

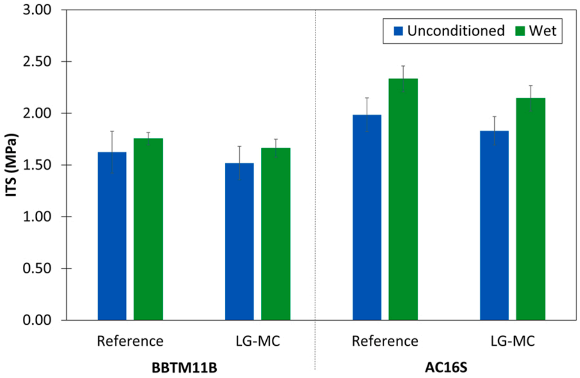

Fig. 6. Average values of indirect tensile strength (ITS) of the reference asphalt mixture and the mixture partially manufactured with LG-MC, types BBTM11B and AC16S, for the different conditions (unconditioned and wet).

Table 5
ANOVA analysis. Null hypothesis: the ITS values of the mixture with the reference filler are similar to those of the mixture with LG-MC. Significance level: 0.05.

Mixture	Condition	Source	Sum of squares	df	Mean square	F	Sig.
BBTM11B	Uncond	Between groups	0.022	1	0.022	0.668	0.445
		Within groups	0.201	6	0.033		
		Total	0.223	7			
	Wet	Between groups	0.017	1	0.017	3.021	0.133
		Within groups	0.034	6	0.006		
		Total	0.051	7			
AC16S	Uncond	Between groups	0.041	1	0.041	1.867	0.230
		Within groups	0.110	5	0.022		
		Total	0.151	6			
	Wet	Between groups	0.071	1	0.071	4.588	0.076
		Within groups	0.092	6	0.015		
		Total	0.163	7			

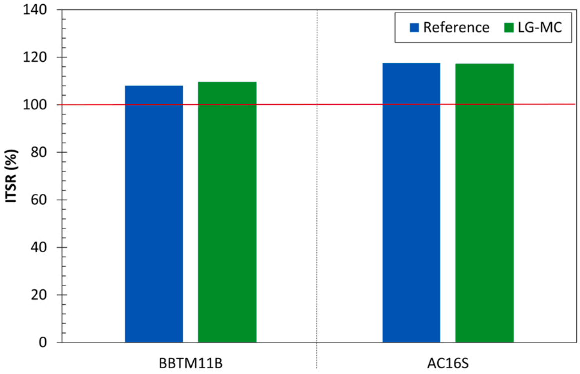

Fig. 7. Indirect tensile strength ratio (ITSR) of the reference asphalt mixture and the mixture partially manufactured with LG-MC, types BBTM11B and AC16S.

used, while these values are lower in the AC16S mixture.

As expected, RT increases with decreasing test temperature and as the mixture ages. This is because of the increase of the mixture stiffness due to the effect of temperature and/or ageing. From the RT results, it is observed that the maximum stiffness that the BBTM11B mixture can reach because of temperature and/or ageing is lower when it is aged. This could be an indication that this type of mixture (a gap-graded one), under extreme conditions (very aged and at low temperatures), could crack sooner than expected.

The average results of the tensile stiffness index, IRT, were obtained from the Fénix test (Fig. 9), obtaining similar conclusions to those observed for the RT results. There are no significant differences between the IRT values of the mixture with the reference filler and the mixture with LG-MC. However, at medium temperatures (20 °C), these values tend to be slightly higher in the mixture with LG-MC.

Table 6
Average values of bulk density and voids content of the specimens tested at Fénix.

Mixture	Condition	Temperature (ºC)	BBTM11B		AC16S	
			Bulk density (g/cm^3)	Voids content (%)	Bulk density (g/cm^3)	Voids content (%)
Reference	Uncond	20	2.107	15.9	2.441	3.0
		5	2.125	15.2	2.439	3.1
		-5	2.118	15.4	2.442	3.0
	LTA	20	2.061	17.7	2.433	3.3
		5	2.062	17.7	2.428	3.5
		-5	2.064	17.6	2.434	3.3
LG-MC	Uncond	20	2.121	16.0	2.421	3.9
		5	2.122	16.0	2.422	3.9
		-5	2.122	16.0	2.423	3.8
	LTA	20	2.093	17.2	2.403	4.6
		5	2.094	17.1	2.403	4.6
		-5	2.096	17.1	2.403	4.6

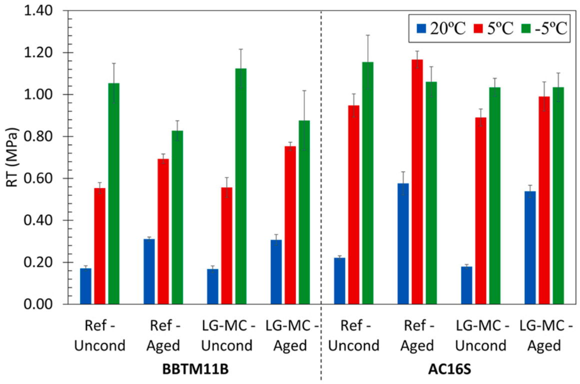

Fig. 8. Average values of the maximum strength (RT) obtained by the Fénix test at the temperatures of 20, 5 and − 5ºC. Results of the reference asphalt mixture and the mixture partially manufactured with LG-MC, types BBTM11B and AC16S, for the different conditions (unconditioned and aged).

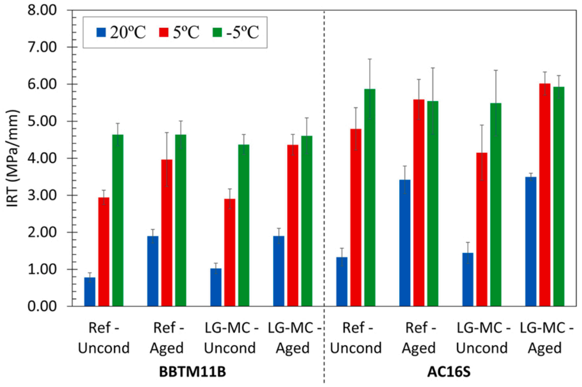

Fig. 9. Average values of the tensile stiffness index (IRT) obtained by the Fénix test at the temperatures of 20, 5 and − 5ºC. Results of the reference asphalt mixture and the mixture partially manufactured with LG-MC, types BBTM11B and AC16S, for the different conditions (unconditioned and aged).

Likewise, it is observed that the BBTM11B mixture presents IRT values lower than those of the AC16S mixture. This is consistent due to the type of binder used in their manufacture, as well as the higher voids content of BBTM11B mixture (Table 6). Obviously, the IRT values increase with decreasing temperature and as the mixture ages. It is observed that, in general, values of IRT at 5 and − 5ºC are similar considering their standard deviation. This is indicating that at 5ºC the mixture has reached it maximum stiffness.

To evaluate how LG-MC influences the ductility of bituminous mixtures against cracking, the toughness displacement (DT) was obtained. Fig. 10 shows the average values of DT for the specimens tested for each condition considered. Results show that at medium temperatures (20ºC), the mixtures with the reference filler present slightly higher ductility values than the mixtures with LG-MC. The particles of calcium carbonate are more rounded than the ones of LG-MC [19]. This could be influencing the cohesion of the filler-bitumen matrix of mixtures made with calcium carbonate presenting higher ductility values. When the mixtures are aged, the DT values are higher when they are partially manufactured with LG-MC compared to those of the reference filler. This indicates that mixtures with LG-MC maintain their ductility to a greater extent after ageing. When the temperature decreases, in general, the mixture with LG-MC presents higher DT values than the one made with the reference filler, which indicates that it is less affected by temperature. In addition, it is observed that the BBTM11B mixture presents a higher ductility than the AC16S one for all cases studied.

The Fénix diagram [30] has been represented in order to analyse the effect of LG-MC on the cracking resistance as a whole, Fig. 11. This diagram allows the graphical analysis of the strength and ductility characteristics of the mixture with temperature. This diagram shows the limits that, according to the authors of the test method experience, separate the fragile, ductile, and high ductility behaviour of the mixture (vertical lines), as well as the low resistance mixtures (horizontal line). In addition, this diagram shows the isotenacity curves for which the RTxDT product remains constant.

For the unconditioned BBTM11B mixture, it is observed that, at medium temperatures, both the mixture made with the reference filler and the one made with LG-MC have very high ductility, presenting the mixture with LG-MC lowest ductility values. However, when the test temperature drops, the mixture with LG-MC maintains a higher ductility. When the mixture ages, the mixture made with LG-MC keeps higher ductility throughout the temperature range, and the ductility range of the aged mixture is between the ductile and very ductile mixtures. When the strengths are compared, it is observed that they are very similar for the same condition studied.

For the case of the AC16S mixture, the characteristic curves depending on the filler used are more similar both for the unconditioned and for the aged condition, and the strength and ductility values are very close, especially at low temperatures. As it ages, the ductility values decrease until they are close to the limit from which the mixture exhibits fragile behaviour. In this case, both strength and ductility are lower in the mixture with LG-MC compared to the mixture with the reference filler.

In summary, by lowering the test temperature for the aged BBTM11B mixture, the one made with LG-MC shows higher ductility values than those of the mixture with the reference filler. This highlights the LG-MC protective effect against the loss of ductility due to ageing and/or the decrease in temperature. Likewise, it is observed that both the aged BBTM11B and the aged AC16S mixtures show a fragile behaviour at low temperatures.

An ANOVA analysis of the RT and DT results was carried out to determine if there are significant differences between the mixture with the reference filler and the mixture partially manufactured with LG-MC. Thus, Table 7 shows the probability of no significant differences between both mixtures with a significance level of 0.05. The results of this analysis show that at 20ºC there are no significant differences in the RT values of the mixture with the reference filler and the mixture with LG-MC, except for the unconditioned AC16S mixture; however, after analysing the results, it is concluded that the differences in RT values are probably due to the voids content which is slightly higher in the LG-MC mixture. Regarding the DT values, there are no significant differences between the BBTM11B mixture with the reference filler and the one partially made with LG-MC. However, for the AC16S mixture, significant differences are observed in the DT values of the mixture with the reference filler compared to the mixture with LG-MC.

At the temperature of 5ºC, no significant differences are observed in the RT values of unconditioned mixtures. When ageing, significant differences in the RT values are observed, which shows the effect of LG-MC on the mixture resistance to ageing. As for the DT values, significant differences are observed when the BBTM11B mixture is unconditioned, and no differences are observed when it is aged. This may be since the BBTM11B mixture has reached its maximum stiffness when it is aged, not obtaining differences in the DT values for the same type of mixture regardless of whether LG-MC was used to make the mixtures. For the AC16S mixture, there are no significant differences in the DT values for the unconditioned mixture, but there are when the mixture is aged. The mixture with LG-MC preserves its ductility to a greater extent.

At − 5ºC, in general, no significant differences are observed in the RT values. At this temperature, the mixture is so stiff that LG-MC does not affect its behaviour. For the DT values, there are only significant differences between the AC16S mixture with the reference filler and the mixture with LG-MC when it is aged. Again, the results are indicating that LG-MC provides a protective effect to the mixture that allows it to maintain its ductility to a greater extent.

The ageing index [31] was obtained to analyse the effect of ageing on the maximum strength and the toughness displacement (Fig. 12). This index was determined using the average values of both parameters. In the case of the maximum strength, values above 1 imply that the RT increases with ageing. From these results, it can be concluded that there is no clear trend of the effect of LG-MC on the ageing of the material. Thus, in general, it is observed that the BBTM11B mixture is similarly affected by ageing regardless of whether LG-MC is used for its manufacturing. In the case of the AC16S mixture, for a temperature of 20ºC, the effect of ageing is greater in the mixture with LG-MC. The opposite effect is observed at 5ºC. At − 5ºC, the effect of ageing is low, especially in the AC16S mix, which is stiffer than the BBTM11B.

In the case of the toughness displacement (DT), the values of the ageing index are below 1. That is, ageing causes a decrease in DT and, therefore, in the ductility of the mixture. The values closer to 1 indicate that the effect of ageing is smaller. For the BBTM11B mixture, the effect of ageing on ductility is less when the mixture is made with LG-MC except for the temperature of 5ºC. At this temperature, the unconditioned mixture does not behave in a brittle manner. However, when combining the effect of ageing with a low temperature, such as 5ºC, the material begins to behave brittle, increasing the effect of ageing when combined with temperature. At very low temperatures, − 5ºC, the mixture behaves very brittle way, so the effect of ageing seems to be reduced as a consequence of the temperature. For the AC16S mixture, the effect of ageing is slightly higher in the mixture with LG-MC at 20ºC. The opposite is observed at lower temperatures. The results of the ageing index once again highlight the protective effect of LG-MC against the fragility of the mixture due to temperature and/or ageing.

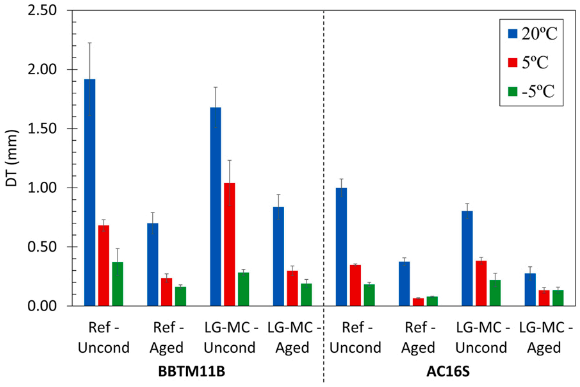

Fig. 10. Average values of the toughness displacement (DT) obtained by the Fénix test at the temperatures of 20, 5 and − 5ºC. Results of the reference asphalt mixture and the mixture partially manufactured with LG-MC, types BBTM11B and AC16S, for the different conditions (unconditioned and aged).

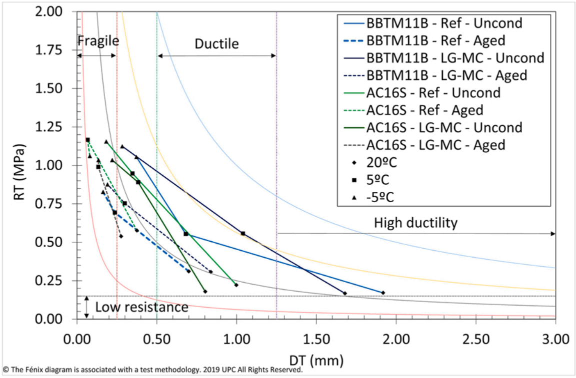

Fig. 11. Fénix diagram. Variation of the maximum strength (RT) with the toughness displacement (DT) of the reference asphalt mixture and the mixture partially manufactured with LG-MC, types BBTM11B and AC16S, for the different conditions (unconditioned and aged) and test temperatures (20, 5 and −5ºC).

Table 7
ANOVA analysis of the RT and DT parameters. Null hypothesis: there are no significant differences in the values of the mixture with the reference filler and the mixture with LG-MC. Significance level: 0.05.

Temperature (ºC)	Mixture	Conditioning	Sig. - RT	Sig. - DT
20	BBTM11B	Uncond	0.727	0.222
		LTA	0.784	0.117
	AC16S	Uncond	0.001	0.007
		LTA	0.270	0.023
5	BBTM11B	Uncond	0.926	0.035
		LTA	0.007	0.060
	AC16S	Uncond	0.169	0.100
		LTA	0.012	0.009
-5	BBTM11B	Uncond	0.329	0.178
		LTA	0.542	0.273
	AC16S	Uncond	0.123	0.254
		LTA	0.613	0.017

3.2.3. *Cántabro test*

The Cántabro test allows to evaluate the cohesion of the aggregate-binder matrix. This property is fundamental since the resistance of the mixture to the efforts of the traffic without disintegrating depend on that. The average values of bulk density and air voids content of the specimens subjected to Cántabro test for the different conditions studied are presented in Table 8.

Fig. 13 shows the average values of Cántabro losses of the BBTM11B mixture for the different conditions evaluated (unconditioned, subjected to moisture damage and long-term aged). It is observed that in the case of the unconditioned mixture, losses tend to be

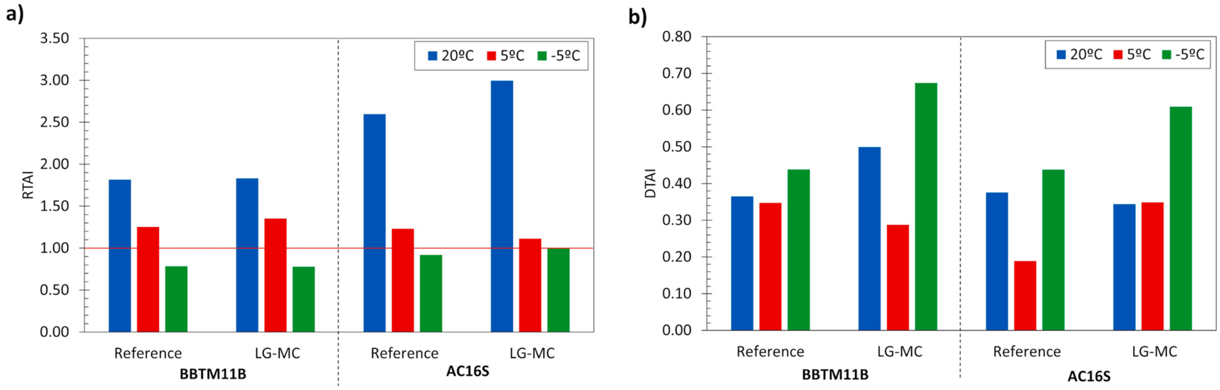

Fig. 12. Ageing index obtained for the parameters: (a) maximum strength and (b) toughness displacement. Results of the reference asphalt mixture and the mixture partially manufactured with LG-MC, types BBTM11B and AC16S, at the test temperatures of 20, 5 and − 5ºC.

slightly lower when the mixture is partially manufactured with LG-MC. By conditioning the mixture, both after immersion and after ageing, these losses increase. Thus, losses tend to be slightly higher in the mixture subjected to moisture damage when it is partially made with LG-MC. However, when the mixture is long-term aged, losses seem to be lower in the mixture with LG-MC. Although slight differences are observed between the mixture with the reference filler and the mixture with LG-MC, in all the study cases, considering the dispersion of the results, it can be shorted that the behaviour of the mixture is similar regardless of whether LG-MC is used or not. This is demonstrated from the ANOVA analysis of Table 9.

From the results obtained, the effect of the bituminous mixture voids content on the percentage of Cántabro losses is observed (Fig. 14). The higher the void content of the mixture for the same conditioning (unconditioned, subjected to moisture damage or aged), the greater the losses are. For the moisture conditioned specimens, it is observed that air voids are slightly higher for the mixture made with LG-MC. This could be the reason why losses are greater for the mixture partially made with LG-MC. However, differences are not significant.

From the analysis of the results, it can be deduced that the mixtures BBTM11B and AC16S partially manufactured with LG-MC present a similar behaviour of the reference one. In the case of ductility at low temperatures or after ageing, the use of LG-MC could have some advantages. However, this analysis is limited to the percentage of LG-MC used compared to the total amount of aggregates needed to manufacture asphalt mixtures.

4. Conclusions

In this research, the use of LG-MC by-product as a partial replacement of the fine fraction of aggregate and as a filler in the manufacture of asphalt mixtures was studied, obtaining the following conclusions:

- The addition of LG-MC did not affect the indirect tensile strength results, regardless of the type of mixture (BBTM11B and AC16S). Values of indirect tensile strength ratio close to 100% were obtained.
- The RT of the unconditioned mixtures evaluated by Fénix test was similar. No significant differences were observed between the mixture with the reference filler and the mixture with LG-MC for the different test temperatures considered (20, 5 and −5ºC) and conditions (unconditioned or aged). Regarding DT values, the mixture with LG-MC tended to preserve ductility to a greater extent at low temperatures as well as when the mixture was aged.
- The cohesion loss resistance of BBTM11B mixture with LG-MC in the Cántabro test can be considered similar to that of the reference.

According to the results, it can be concluded that LG-MC offers the mixture some protective effect in its resistance to cracking. These results show the suitability of this by-product as a partial substitute for the fine fraction of aggregates, as well as for the total amount of filler in asphalt mixtures manufacturing for road pavements. Thus, the use of this by-product would contribute to reduce the use of natural aggregates, fighting against the depletion of non-renewable resources, as well as to reduce the waste that ends in landfills,

Table 8
Average values of bulk density and voids content of the BBTM11B mix specimens used for the Cántabro test.

Condition	Reference		LG-MC	
	Bulk density (g/cm^3)	Voids content (%)	Bulk density (g/cm^3)	Voids content (%)
Uncond	2.099	16.2	2.127	15.8
Wet	2.119	15.4	2.127	15.8
LTA	2.063	17.6	2.093	17.1

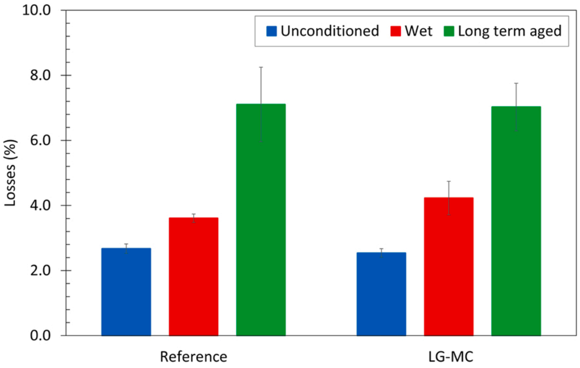

Fig. 13. Average values of Cántabro losses of the reference asphalt mixture and the mixture partially manufactured with LG-MC, type BBTM11B, for the different conditions (unconditioned, wet and long-term aged).

Table 9
ANOVA analysis. Null hypothesis: the behaviour of the mixture with LG-MC against abrasive wear is similar to that of the mixture with the reference filler. Significance level: 0.05.

Condition	Source	Sum of squares	df	Mean square	F	Sig.
Uncond	Between groups	0.035	1	0.035	1.379	0.285
	Within groups	0.153	6	0.025		
	Total	0.188	7			
Wet	Between groups	0.773	1	0.773	4.115	0.089
	Within groups	1.126	6	0.188		
	Total	1.899	7			
LTA	Between groups	0.013	1	0.013	0.010	0.923
	Within groups	7.370	6	1.228		
	Total	7.383	7			

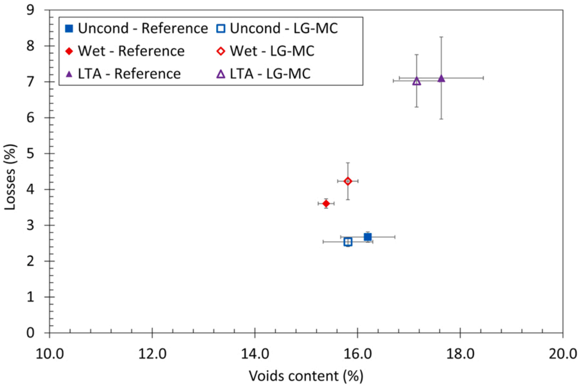

Fig. 14. Percentage of losses against voids content of the reference asphalt mixture and the mixture partially manufactured with LG-MC, type BBTM11B, for the different study conditions (unconditioned, wet and long-term aged).

promoting circular economy.

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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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