

Preliminary mechanical and environmental assessment of bituminous mixtures including a graphene-based compound

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ABSTRACT

The constant increase in the performance request and the need to preserve the environment for future generations drive to the development of sustainable technologies and materials. Within this framework, extending pavement service life, reducing maintenance frequency, and limiting the consumption of virgin raw materials are key targets. Also, the reuse of reclaimed and end-of-life materials can contribute to these objectives, provided that the resulting mixtures maintain adequate mechanical and functional performance. In this perspective, this study investigates an innovative graphene-based additive incorporating selected recycled hard plastics for asphalt wearing courses. For this purpose, different mixtures have been produced with the additive via dry process and subjected to both functional and mechanical characterization, in order to compare the modified solutions with conventional and high-performance reference mixtures. The test results indicate that the use of a compound based on graphene and recycled hard plastics can improve selected mechanical performance indicators of the mixtures, especially indirect tensile strength, stiffness and rutting resistance. The investigated surface indicators remained within the relevant specification limits, although some variations with respect to the reference mixtures were observed. For a more robust evaluation of the experimental mixtures, they were also comparatively studied in terms of environmental impact using the Life Cycle Assessment (LCA) methodology. Within the adopted cradle-to-gate boundary, the results indicate reductions in energy consumption and related emissions for the modified mixtures. Overall, the findings support the technical feasibility of the proposed additive for the investigated asphalt mixtures, while conclusions regarding long-term durability, service-life extension and whole-life sustainability require further validation.

1. Introduction

Sustainability has become one of the principal goals of the last decade; in fact, nowadays human activities are directed to protect the environment and to preserve the current resources on earth for future generations.

At the same time, in transport infrastructure engineering, increasing traffic volume and axle loads require constant improvement in the performance of the various materials that compose a road pavement.

Consequently, current research aims to develop improved road technologies and materials able to extend pavement service life and reduce maintenance needs, while improving the circular use of materials derived from by-products or end-of-life materials. These include

strategies that promote the rejuvenation and reuse of asphalt obtained from the demolition or milling of existing layers, as well as the recycling of materials at the end of their service life, minimizing energy consumption and related emissions. Road paving technologies are constantly evolving not only to achieve better levels of performance and durability, but also to pursue goals such as greater environmental effectiveness, improved user safety, reduced pollution, lower energy consumption and lower architectural impact [1,2]. From this point of view, a reliable method already consolidated in the literature is the use of additives such as polymers or fibres in asphalt mixtures. Polymeric compounds, in particular, can enhance mixture performance under different climatic conditions and loading regimes [3]. In several cases, these additives may also contribute to lower maintenance demand and,

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consequently, to reduced life-cycle impacts [4–6]. Despite these interesting advancements, research is still ongoing to evaluate other materials able to ensure both performance and environmental benefits.

A relevant alternative, in this context, could be represented by graphite-based additives. Graphite is a layered material whose structure is formed by carbon atoms set together on the same level by covalent bonds. Deriving from the processing of graphite, graphene is a monolayer of carbon atoms arranged in a honeycomb structure. Wallace theoretically studied the electronic structure of a graphite monolayer in 1947, while graphene was experimentally isolated in 2004 [7,8].

The main characteristics of graphene are the small thickness, the very high resistance and flexibility, the excellent heat and electricity conduction, the huge light absorption and the extremely high specific area. These features make it excellent for products in several high technological fields such as electronics, aeronautics, medicine, astronautics, textile, sport, environment [1], and, more recently, for civil engineering materials, including asphalt binders and mixtures.

Graphene base materials are GOs (graphene oxide nanoplatelets), GO (graphene oxide: it is prepared by oxidation of graphite powder in laboratory), GNP (graphene), GNPs (graphene nanoplatelets). Graphene and GO are two of the most promising materials due to their high specific surface area and excellent electrical and physical properties. Earlier applications were limited by high production cost and dispersion issues, whereas recent advances have encouraged further studies [7].

In the road sector, the use of graphene-based nanomaterials to modify the asphalt mixture and enhance its performance has become an area of growing interest: promising effects have been reported on self-healing ability, conductivity, viscosity, mechanical performance, compaction performance, and high temperature performance. However, some studies also report potential drawbacks related to low temperature performance and phase separation [9].

A recent study also reported that mixing binder with GO enhances resilient and moisture stability and improves the fracture resistance of the asphalt mixture [10]. GO has potential advantages in improving pavement performance, including ageing resistance, UV ageing resistance and thermo-oxidative ageing resistance of asphalt [11,12]. It was also demonstrated that thermodynamic parameters and intrinsic healing capacity of GO-modified asphalt binders increase after short-term ageing in the laboratory [13]. Furthermore, GO can improve anti-ageing properties, high-temperature stability and temperature sensitivity due to increased viscosity [14]. Another study also reported that the addition of GNPs can improve pavement performance in terms of ITS, water sensitivity, stiffness modulus and fatigue resistance [15].

Russo et al. [16] also highlighted the benefits of dry application, reporting improvements in stiffness modulus, indirect tensile strength, compaction behaviour, and production-stage energy demand.

More recent studies further confirmed the interest in graphene-related and graphene-polymeric asphalt solutions, including reviews of graphene materials in asphalt mixtures, field-oriented case studies on graphene-enhanced polymeric compounds, and investigations on ageing behaviour of GO/SBS-modified binders [17–20]. These studies generally report promising effects on high-temperature behaviour, ageing resistance, moisture-related properties or field performance; however, they also underline that dosage, dispersion, binder chemistry and preparation protocol strongly affect the final response of the material. Furthermore, graphene-based additives have also been studied for evaluating their potential in improving asphalt energy efficiency and developing interesting emerging technologies, such as the so-called hydronic asphalt pavement (HAP), with energy-harvesting properties [21,22]. A recent study highlights the feasibility of de-icing using appropriate quantity of graphene because its addition can endow bituminous material with electrical conductivity [23].

Based on this background, the scientific gap addressed in this paper concerns the laboratory performance and production-stage environmental assessment of dense and porous wearing-course mixtures incorporating a commercial graphene-based additive containing

recycled hard plastics, designed under Italian specification conditions.

Accordingly, in this paper, the potential mechanical benefits of an innovative asphalt mixture, modified through dry addition with a graphene-based additive containing recycled waste plastics, are preliminarily investigated [6]. The objective is to evaluate whether such modification can improve mechanical performance while maintaining adequate surface functionality and reducing “cradle-to-gate” environmental burdens [24,25]. To this end, the proposed mixtures were compared with conventional and high-performance reference solutions through laboratory testing and life cycle assessment (LCA). It is opportunistically clarified that the contribution of the work is therefore framed as a comparison of reference and modified mixtures, rather than as an attempt to isolate the specific effect of graphene from that of the recycled polymeric fraction.

2. Materials and methods

2.1. Materials

2.1.1. Mineral aggregates

The aggregates used in this research work were crushed limestone from a local quarry. Figs. 1 and 2 show the aggregates composition for “dense” and “porous” surface courses, while Table 1 provides aggregates physical and mechanical properties.

2.1.2. Bituminous binders

Three different types of bitumen are used in this study: two neat bitumen with different penetration grade, and one polymer modified bitumen. More specifically, a 50/70 penetration grade bitumen was used for both dense and porous surface course mixes, while a 35/50 penetration grade bitumen for the latter mix only. Additionally, the polymer modified bitumen, named “hard bitumen” (HD), modified with SBS, was selected for both the mixtures. All binders were provided by a local company. The general properties of all bitumens involved in this study are detailed in Table 2.

2.1.3. Additives

The graphene-based additive used in this study was provided by an Italian company. As anticipated, the product contains not only graphene but also a selected fraction of recycled hard polymers. The functional base consists of various additives, but the physico-chemical composition is confidential for industrial reasons. The graphene used is composed of highly pure graphene nanoplatelets.

The additive (Fig. 3), hereafter referred to as GP, is produced through an industrial process intended to valorise recycled polymeric fractions. Preliminary studies, both in laboratory and in situ, have proved a potential of this additive as a mixture modifier, in terms of mechanical performance and durability [15].

The main properties of the additive, as reported in the technical data sheet provided by the manufacturer, are summarised in Table 3.

The confidential nature of the complete formulation represents a limitation in terms of full reproducibility. For this reason, the present study evaluates the additive as a whole commercial composite modifier, and the results should not be interpreted as the isolated effect of graphene alone. To improve transparency, all non-confidential information available from the manufacturer is reported, and the dry-process mixing protocol adopted in the laboratory is described in the following section.

2.1.4. Experimental plan

As schematically shown in Fig. 4, the experimental plan included standard laboratory tests on aggregates, filler, binders, and asphalt mixtures. The first phase of the investigation consisted of the systematic sampling and physico-chemical characterisation of all constituent materials.

Then, two mixtures commonly employed in surface courses were investigated in this study: a dense-graded mixture and an open-graded

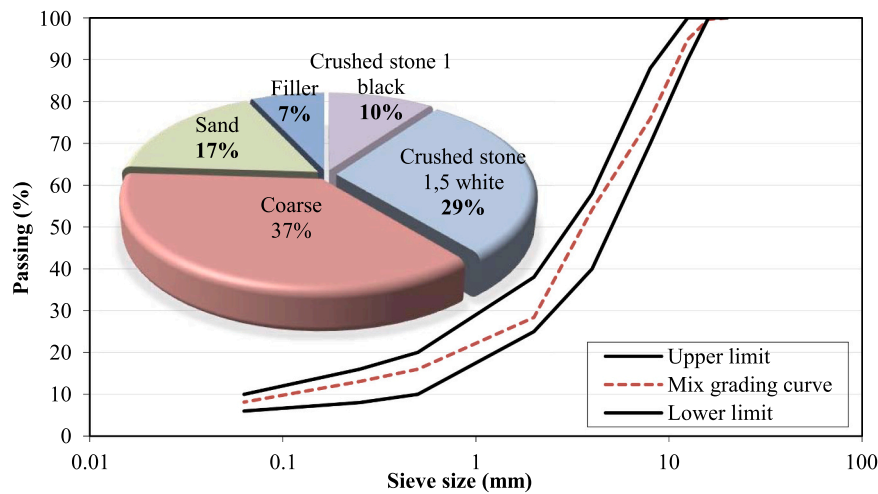


Fig. 1. Dense surface course mix grading curve.

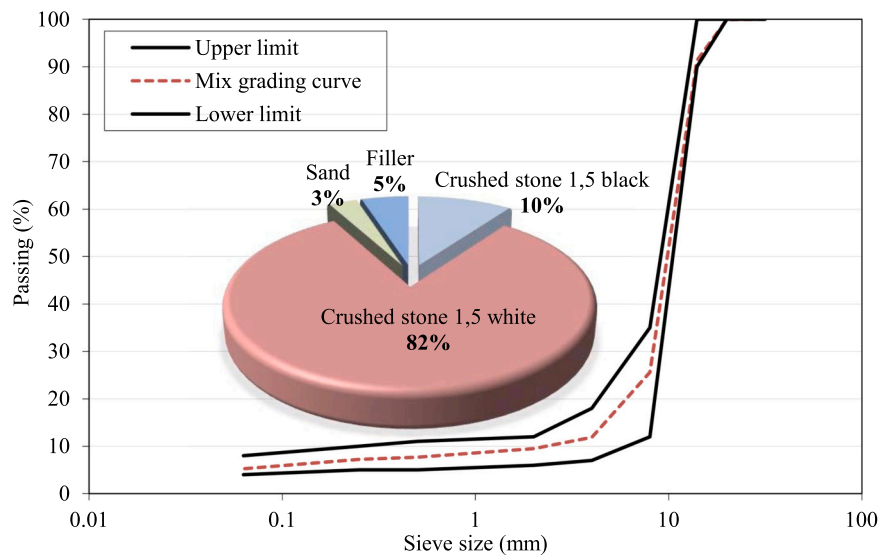


Fig. 2. Porous surface course mix grading curve.

Table 1

Physical and mechanical characteristics of the aggregates used in this study.

Characteristics	Unit	Crushed stone 1,5 white	Crushed stone 1,5 black	Crushed stone 1 black	Coarse	Sand	Filler	Standard
Flattening index	%	5			5			EN 933-3:2012
Real specific weight (γ_r)	g/cm ³						2.848	EN 1097-7:2008
Apparent specific weight (γ_a)	g/cm ³	2.844	3.568	3.444	2.848	2.852		EN 1097-6:2013
Los Angeles abrasion	%	17	10	15	20			EN 1097-2:2020
Frost susceptibility	%	0.5	0.5		0.6			CNR 80/80
Sand equivalent	%					85		EN 933-8:2012

mixture, named as "dense surface course" and "porous surface course", respectively, in accordance with Italian road specification [26].

Both the dense wearing course and a porous wearing course were produced with a polymer-modified bitumen (PMB HD) to ensure high thermal, mechanical and functional performance in line with traffic and safety conditions. The GP-modified mixtures were instead produced with conventional neat bitumen and the additive, while maintaining the same aggregate skeleton. This comparison made it possible to assess whether the graphene-based additive could compensate, for the use of a less performing binder.

For the mixtures with neat bitumen, according to UNI EN 12697-35,

the mixing temperature was set at 160°C, whereas for all the other mixtures containing the polymer modified bitumen or additive the mixing temperature was set at 175°C, considered as a maximum to prevent excessive ageing of the bituminous binder.

The additive content was set at 6% by mass of the binder; that is, 6% of the total binder mass was replaced with the graphene-based additive. According to the manufacturer guidelines, GP was added into the mixture after the bitumen and before the filler; then, the mixture of aggregates and additive was conditioned in the oven at mixing temperature, for approximately 30 min to promote polymer softening, and for about 2 h before compaction with the gyratory compactor.

Table 2
Characteristics of the bitumens used in this study.

Characteristics	Unit	50/70	35/50	HD	Standard
Specific weight at 25°C	g/cm ³	1.03	1.03	1.04	EN 3838:2005
Penetration at 25°C	dmm	51	36	50.8	EN 1426:2015
Ring and ball softening point	°C	54.2	54.5	88.5	EN 1427:2015
Fraas breaking point	°C	-7			EN 12593:2015
Ductility at 25°C	cm	> 100			EN 13398:2018
Elastic return	%			96	EN 13398:2018
Viscosity at 160°C After RTFOT:	Pa•s	0.13	0.18	0.99	EN 13302:2018
					EN 12607-1:2015
Retained Penetration at 25°C	%	64.7		40	EN 1426:2015
Ring and ball softening point	°C	57.2		91.5	EN 1427:2015
Viscosity at 160°C	Pa•s			1.07	EN 13302:2018



Fig. 3. The graphene-based additive used in this study.

Table 3
Characteristics of the graphene-based additive used in this study.

Characteristics	Unit	Additive
Aspect	-	Granules
Colour	-	Black
Apparent density at 25°C	g/cm ³	0.4÷0.6
Softening point	°C	160÷180
Melt flow index at 190°C/5 kg	g/10'	4÷10

The selected dosage was adopted according to the manufacturer guidelines and preliminary feasibility considerations related to dry-process workability and volumetric compliance. This value should therefore be considered a representative dosage for this experimental comparison, not an optimum content. A dedicated dosage-optimisation study, including lower and higher additive contents and additional durability indicators, is required to identify the optimum formulation.

For clarity, the designation of the mixtures is provided in Table 4.

Because the study was organised in sequential stages, not all mixtures were subjected to the full experimental programme. All mixtures listed in Table 4 were included in the volumetric study and in the indirect tensile strength screening, which were used to verify specification compliance and to identify the most promising formulations. Wheel-tracking tests were then carried out on the selected design mixtures for each family in order to compare rutting resistance under severe loading, whereas stiffness modulus and surface-characterisation tests were limited to the final optimised mixtures, i.e., the reference high-performance solutions and the corresponding GP-modified alternatives.

This staged approach was adopted to keep the campaign experimentally consistent while concentrating the more time-consuming tests

on the mixtures considered most representative for final comparison.

Some binder-level tests indicated in the experimental framework, such as DSR and storage-stability checks, were used as preliminary or supporting characterisation activities and are not discussed in detail in the present manuscript, which focuses on mixture-level performance and cradle-to-gate LCA. This has been explicitly considered in the interpretation of the results and in the conclusions.

2.2. Methods

2.2.1. Mixture volumetric study

The mix design was developed in compliance with current specifications for porous and dense asphalt mixtures.

The optimum binder content (OBC) was determined through volumetric analysis using a gyratory compactor. Additionally, in compliance with EN 12697-31, the compatibility of the mixtures was assessed; this test, in fact, as known, provides essential insights into the workability of the mixture and its potential rutting resistance [6].

The gyratory compactor test was first carried out on cylindrical specimens (D=100 mm) of the conventional mixtures (without additive) only, by varying the percentages of bitumen; after OBC of the reference mixtures had been identified, this test was also performed on the corresponding GP mixtures, at the selected additive content. Italian Specifications define acceptance ranges for air voids at three compaction levels: N₁, N₂, N₃ [26].

Volumetric properties were determined in accordance with the requirements of EN 12697-8. Finally, the maximum specific gravity of the mixture (γ_t) was calculated in accordance with EN 12697-5, while the bulk specific gravity (γ_{app}) was determined according to EN 12697-6.

2.2.2. Mechanical performance

To determine the OBC and to compare mixture strength, the indirect tensile strength test was performed at a temperature of 25 °C. This test, carried out according to EN 12697-23, allowed the assessment of the indirect tensile strength (ITS) and the indirect tensile coefficient (CTI). ITS and CTI can respectively be calculated through Eq. (1) and Eq. (2).

$$ITS = 2 P / (\pi h D) \quad (1)$$

$$CTI = (\pi D ITS) / (2 d_v) \quad (2)$$

where P represents the peak value of the applied vertical load in N, h and D are respectively the mean thickness and the diameter of the cylindrical specimen - expressed in mm - and d_v is the vertical deformation at failure, in mm.

Moreover, in order to evaluate the rutting resistance of the investigated mixtures, wheel tracking tests in air were carried out according to EN 12697-22, Method B. Two slabs were produced for each mixture with the dimensions of 305 mm × 305 mm × 50 mm; and the reported result corresponds to the average of the two tests. The tests were performed at a temperature of 60°C up to 20000 load cycles or to a maximum deformation of 20 mm, at a frequency of 0.44 Hz and a load of 700 N.

Lastly, only the optimized mixtures identified after this testing phase were studied in terms of stiffness modulus by means of the indirect tensile test. Tests were performed on cylindrical specimens (D=150 mm and h=60 mm), produced by gyratory compactor at the target air void content, v , set equal to 4.5% for the dense surface course mix and 26% for the porous surface course mix, at 0, 10, 15, 20 and 30°C, according to EN 12697-26, annex C. This standard prescribes one fixed pulse repetition time (3 s) and suggests a rise time of 124 ms. The stiffness modulus was determined through Eq. (3).

The reported values are average values of the available laboratory replicates (at least three valid replicates per test), as specified in the pertinent standard.

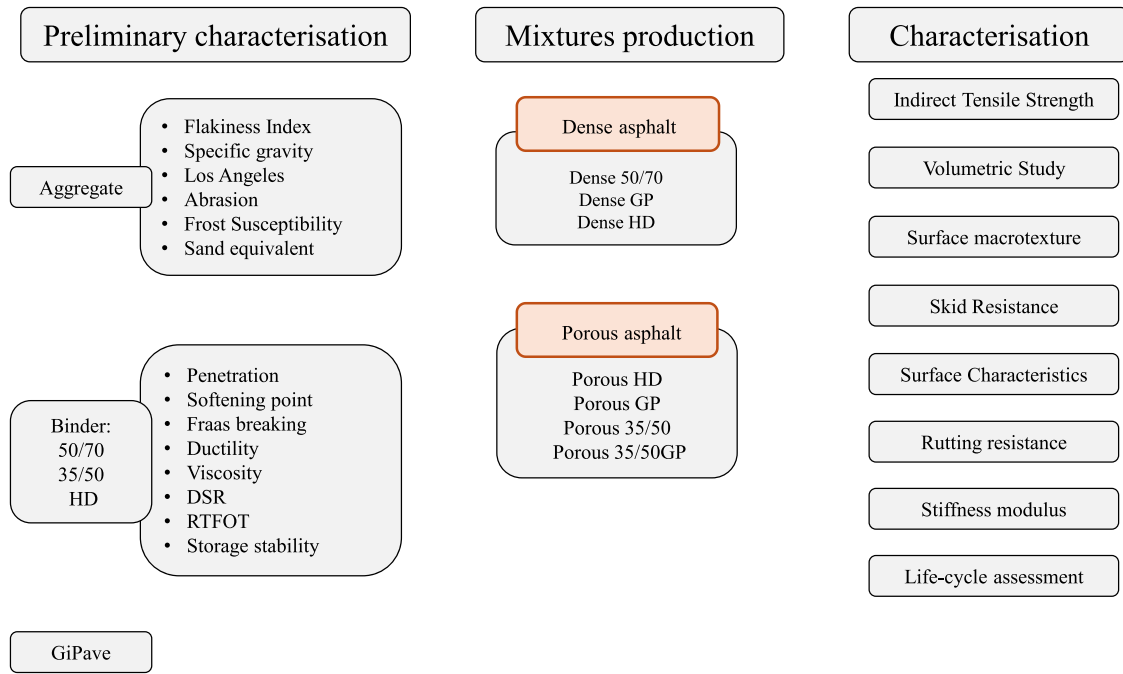


Fig. 4. Flow chart of the experimental plan.

Table 4

Mixture designations.

Name	Bitumen	Additive	Surface-Course
Dense 50/70	50/70 penetration grade	No	A surface course
Dense GP	50/70 penetration grade	Yes	A surface course
Dense HD	Hard Bitumen	No	A surface course
Porous HD	Hard Bitumen	No	Porous surface course
Porous GP	50/70 penetration grade	Yes	Porous surface course
Porous 35/50	35/50 penetration grade	No	Porous surface course
Porous 35/50GP	35/50 penetration grade	Yes	Porous surface course

$$S_m = \frac{[F(\nu + 0.27)]}{(z h)} \quad (3)$$

where S_m is the stiffness modulus in MPa, F is the peak vertical load in N, ν the Poisson's ratio, z the amplitude of the horizontal deformation during the load cycle, in mm, and h the average specimen thickness, in mm.

2.2.3. Surface characteristics

The optimized mixtures were also studied in terms of surface characteristics by means of a contactless laser profilometer based on conoscopic holography [27,28]: this test is useful for evaluating texture aggregate indicators and spectral indicators, whose profiles are disaggregated through Fourier analysis, as well as macrotexture using the volumetric method, skid resistance using the British Pendulum, and drainability.

The tests were carried out on slabs produced for each mixture with dimensions and features analogous to those used for rutting resistance tests (305 × 305 × 50 mm, $\nu = 4.5\%$ for dense surface course and 26% for porous surface course).

In particular, for each sample, the profilometer acquired seven parallel profiles, with a length of about 300 mm. This allowed to obtain a linear profile with a total length of at least 2 m for each mixture, ensuring a good representativeness for the spectral analysis.

The texture data were post-processed through the software U-Pave-Tex, developed for the University of Calabria within an Italian project of relevant national interest (PRIN) "USR342 - Urban safety, Sustainability,

and Resilience: 3 paving solutions, 4 sets of modules, 2 platforms". The software returns the classical roughness parameters (MPD, ETD, R_a , R_u , R_z , R_{max} , RMS); statistical descriptors such as Skewness and Kurtosis, Bearing-Abbot curve parameters, and the spectral indicator Texture Level (LTX). The detailed descriptions of the indicators can be found in [28,29].

2.2.4. Life-cycle assessment – "cradle to gate"

To complete the comparison, a cradle-to-gate LCA was carried out on the investigated mixtures in order to quantify the environmental impacts associated with the products and processes involved. The analysis was performed in accordance with ISO 14040 and ISO 14044 standards, with background datasets implemented in SimaPro 9.4.0.2 software, for unitary impact assessment.

Although LCA is generally intended to cover the full life cycle, the present study focuses on the production stage because this is the phase directly affected by the proposed material substitution [24,25]. The assessment therefore includes goal and scope definition, life cycle inventory, impact assessment, and interpretation:

- Goal and scope: the study objectives, the functional units of the analysed system, and the boundary conditions are assessed;
- Inventory analysis: a complete inventory of the input and output flows related to each operation is assessed. An inventory of the resources used and of the emissions associated with the functional units is also carried out.
- Impact assessment: the potential impact arising from the elementary flows is assessed, and the various impact categories and associated quantities are assigned.
- Interpretation: it is the final stage, in which the results are evaluated in relation to the objectives and the defined scope, in order to carry out the necessary assessments and implement appropriate corrective actions for the process.

Furthermore, the reference unit adopted in this study, according to the standards, is represented by the declared unit (DU), consisting of 1 metric ton of HMA suitable for surface courses, as described in previous sections.

3. Result and discussion

3.1. Mixture volumetric study

This section presents the results of the volumetric study of the mixtures compacted by gyratory compactor at the determined OBC. For brevity, only the final design solutions are discussed here, whereas the interpretation focuses on their volumetric response and implications for mixture stability. In accordance with the staged experimental plan described in Section 2, the volumetric screening and ITS comparison involved the full set of mixtures, whereas the subsequent performance analyses were restricted to the selected benchmark and GP-modified solutions. The main outcomes are reported in Tables 5 and 6 and are discussed in terms of the volumetric and mechanical properties. In accordance with the reference standard, the values reported in the table represent the average of measurements obtained from three valid replicate specimens. The corresponding standard deviation, which describes the variability of the replicate measurements around their mean value, is also reported. The requirements are respected for bitumen content equal to 5.5% for dense surface course mix (Table 5) and bitumen contents between 4.9% and 5.1% for porous surface course mix.

Moreover, a measure of the internal stability of the mixture, the shear ratio, σ , during the compaction was registered and the related values are reported in Figs. 5 and 6. An increasing trend of this parameter in the initial stage of compaction (in the first 50 gyrations) and stabilization beyond the maximum value of N , together with fulfilment of the volumetric requirements, voids in mineral aggregate (VMA) and voids filled with asphalt (VFA), suggest a correct formulation and, thus, good mixture stability under the investigated laboratory conditions. As can be seen from the charts provided in Fig. 5 and Fig. 6, the selected mixtures confirmed this hypothesis, showing substantially constant shear-ratio values after the initial compaction stage.

3.2. Mechanical performance

The cylindrical specimens produced by means of the gyratory compactor (compacted to a number of gyrations equal to N_3) were subjected to increasing load along the diametral plane for the indirect tensile strength test. The compression determines the indirect tensile stress on it up to failure. In accordance with UNI EN 12697–23, the results shown in Figs. 7 and 8 represent the mean values obtained from three valid replicate specimens; error bars are included for clarity. The error bars represent one standard deviation and are provided as a measure of the variability among replicates. Given the limited number of specimens required by the reference test procedure, the statistical analysis was intended to describe test repeatability rather than to support broader inferential conclusions. The values of these parameters for the selected mixtures are reported respectively in Fig. 7 and Fig. 8; the requirements considered in the Italian specifications [26] are listed in Table 7.

As can be seen, the GP-modified mixtures exhibited consistently higher ITS and CTI values than the corresponding mixtures without additive. This trend indicates a consistent enhancement of tensile performance associated with the incorporation of the graphene additive. Specifically, mixtures containing GP exhibit markedly higher ITS values compared to their corresponding reference mixtures, thereby indicating

a favourable effect of the additive across the investigated mixture typologies. However, the results should be interpreted as comparative laboratory evidence and not as a direct surrogate for field cracking performance. Analogously, CTI values for GP mixtures are higher than for conventional mixtures.

Further tests were performed in terms of rutting resistance. In detail, Fig. 9 and Fig. 10 show the resistance to permanent deformation of the dense and porous surface mixtures, respectively Lower rut depth at a given number of load cycles indicates higher resistance to permanent deformation at high temperatures. The reported results represent the mean values obtained from two valid replicates, which satisfied the repeatability limits specified in the relevant reference standard. As can be seen in Fig. 9, at the same traffic volumes, rut depth values for the mixture with graphene-based additive are approximately 71% lower than the reference dense mixture produced with 50/70 bitumen at the same traffic level, for dense surface course mix. Furthermore, these values are comparable with those of the mixture produced with hard bitumen.

For the porous surface course mix, the mixtures with graphene-based additive sustained about 77% more load cycles than the corresponding unmodified mixture at comparable rut depth, as proved in Fig. 10, thus confirming a marked improvement in high-temperature stability.

Additionally, stiffness modulus values are reported in Fig. 11 and Fig. 12, for the dense and porous surface-course mixtures, respectively. As can be seen, in both cases the GP-modified mixtures showed higher stiffness than the corresponding reference mixtures without additive, throughout the investigated temperature range.

The stiffness modulus tests are consistent with the mechanical trends observed in the ITS and wheel-tracking tests. The observed improvements may result from the combined effects of the graphene-based additive and the hard recycled polymer fraction, which can increase mixture stiffness, strengthen the bituminous mastic, and improve resistance to permanent deformation. Nevertheless, a higher stiffness should be interpreted together with the expected service conditions, because possible trade-offs at low temperature or under repeated fatigue loading were not directly investigated in the present study.

Since fatigue resistance, moisture susceptibility, low-temperature cracking and ageing performance were not assessed, the observed increase in stiffness and rutting resistance should not be directly translated into long-term durability or service-life extension. These aspects require dedicated tests before practical implementation can be fully evaluated.

3.3. Surface characteristics

Tables 8 and 9 show the results of the profilometric measurement of the surface texture parameters of the considered asphalt mixtures.

The asphalt mixtures with the highest air void content, Porous Asphalt type, have the highest values of the geometrically relevant texture parameters, see Table 8, which strongly reflect the internal porosity on the surfaces. This result is also evident for the measurements with the volumetric method. Focusing on dense asphalts, it can be seen that for the modified mixes (Dense 50/70 GP and Dense HD) there is an average decrease of about 20% in texture depth (MPDi, ETDi, Ra, Ru, and Rz), if compared with the corresponding conventional dense mixture (Dense 50/70). This difference becomes smaller for RMS (about 8%) and is negligible for the Rmax parameter, which reflects the

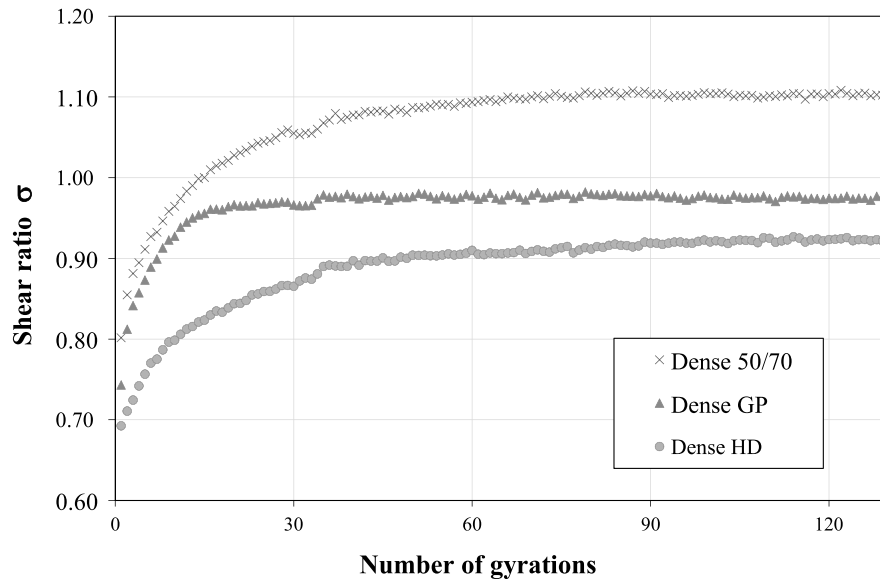
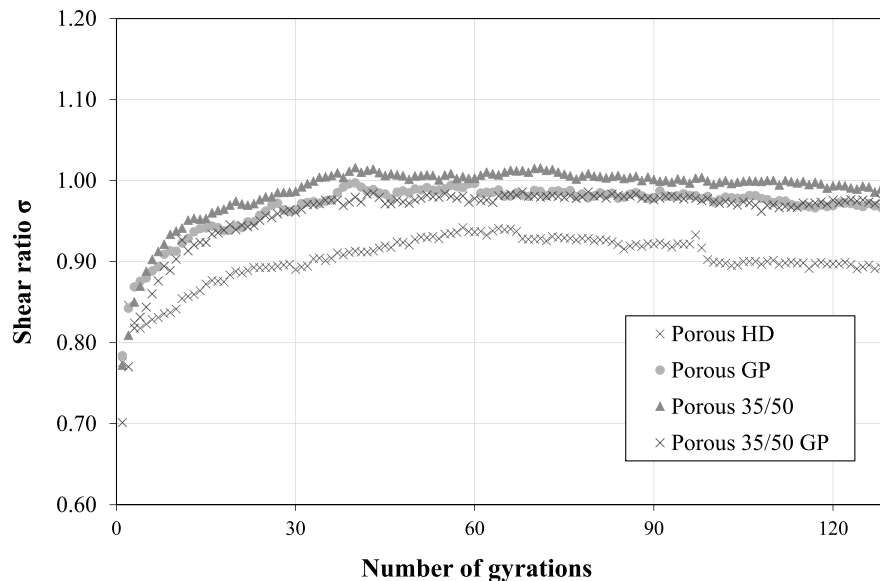
Table 5

Average values of the air voids of the specimens at $N_1=10$, $N_2=120$ and $N_3=210$ gyrations for Dense surface course mix.

Mixtures	% b	Air voids [%]					
		N_1	Standard deviation	N_2	Standard deviation	N_3	Standard deviation
Requirements	4.5÷6.1	11÷15		3÷6		≥ 2	
Dense 50/70	5.5	14.5	±0.32	4.4	±0.76	2.8	±0.78
Dense GP	5.5	14.6	±0.35	5.3	±0.37	3.9	±0.45
Dense HD	5.5	14.4	±0.50	3.9	±0.45	2.6	±0.40

Table 6Average values of the air voids of the specimens at $N_1=10$, $N_2=50$ and $N_3=130$ gyrations for Porous surface course mix.

Mixtures	% b	Air voids [%]					
		N_1	Standard deviation	N_2	Standard deviation	N_3	Standard deviation
Requirements	4.8÷5.8	≥ 28		≥ 22		≥ 20	
Porous HD	5.1	31.2	± 0.65	26.1	± 0.50	23.2	± 0.66
Porous GP	4.9	32.4	± 0.63	26.9	± 0.72	23.9	± 0.62
Porous 35/50	5.1	37.3	± 1.70	32.3	± 1.80	29.7	± 2.10
Porous 35/50 GP	4.9	33.2	± 0.58	26.7	± 0.80	23.2	± 1.06

**Fig. 5.** Shear ratio of dense surface course mix.**Fig. 6.** Shear ratio of porous surface course mix.

maximum peak-to-valley vertical distance along the profile.

For porous asphalts, the differences between modified mixes (Porous 50/70 GP, Porous 35/50, and Porous 35/50 GP) and the reference mix (Porous HD) were modest, on average, less than 5%.

For measurements with the volumetric method, for each type of mixture (dense or porous) evident differences emerged (deviations generally below 5%) with the corresponding control mixtures, probably

due to the intrinsic uncertainty of measurement by the volumetric method.

Kurtosis (R_{ku}) and Skewness (R_{sk}), see Table 9, are parameters that characterize the shape of the probability densities of the texture profile heights [28] [30].

Kurtosis (R_{ku}) describes the probability of flattening the profile. Non-Gaussian surfaces with relatively flat peaks and valleys are

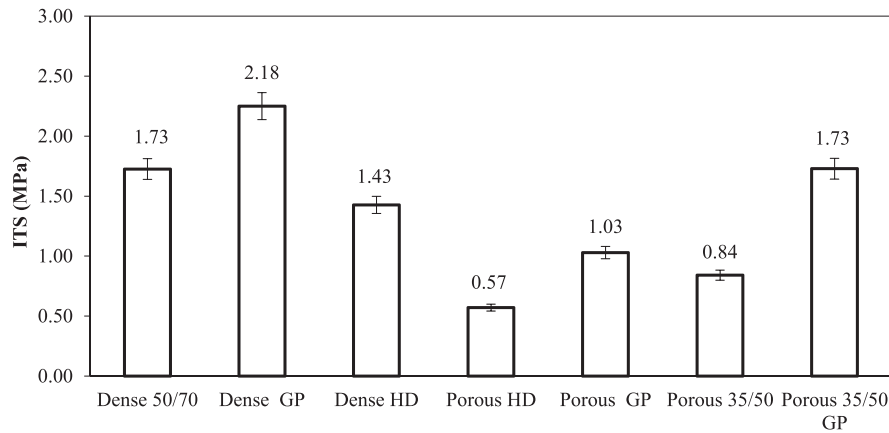


Fig. 7. ITS values of the selected mixtures.

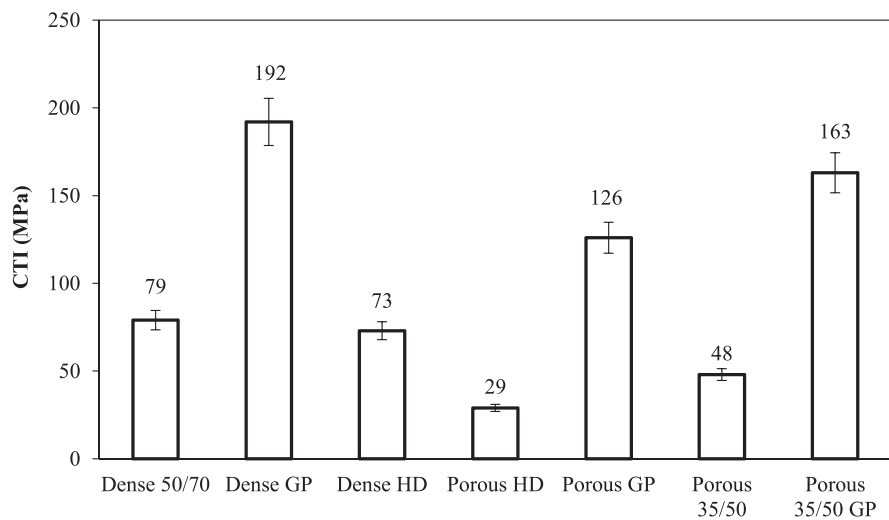


Fig. 8. CTI values of the selected mixtures.

Table 7
Italian ANAS specification standards.

Mixture	Dense 50/70	Dense HD	Porous HD
ITS (MPa)	0.72 – 1.40	0.95 – 1.70	0.36 – 0.70
CTI (MPa)	≥ 60	≥ 70	≥ 30

reflected in a kurtosis less than 3 (platykurtic), while a kurtosis value above 3 (leptokurtic) indicates sharp peaks and valleys [31]. In particular, only the Dense 50/70 GP mixture has an Rku value slightly above 3, while the rest of the mixes (Dense and Porous) have an Rku value of less than 3.

Skewness (Rsk) evaluates the symmetry of the texture profile with the average line of heights. Rsk is sensitive to occasional deep valleys or high peaks because it measures the symmetry of the profile distribution over its midline. A symmetric distribution is reflected in zero skewness. Predominant profiles at peaks above a flatter average are reflected in positive skewness, while negative skewness refers to profiles prevalent in deep valleys [29]. In Table 9, for all mixes (Dense and Porous), $Rsk < 0$ indicates that the peak of the texture profile is smooth, wide, and flat, while the trough of texture is sharp and narrow, which shows negative texture with negative skew distribution [31].

All of the mixtures (Dense and Porous) obtain relatively high shape factors (g-factor): from 71.8 to 76.5. Although asphalt surface layers with higher shape factors have better noise-reducing properties, this is

not the case with porous asphalt, because its noise-reducing mechanism is not based on the surface texture shape, but on the noise absorption by the large connected air voids in the structure of the layer. In fact, generally, this texture parameter is not used for the evaluation of noise-reducing mechanism for porous asphalt mixtures [32].

Generally, in the Abbott curve analysis are fixed the heights of the profile are above or below which a certain portion of peaks or valleys is located, respectively. In road texture surface analysis, usually chose the value above which 10% of the profile peaks lay and the valley beneath which 20% of the valleys were included (80% of the whole profile) [33]. These portions, peaks and valleys, are given by Rpk (peaks) and Rvk (valleys), and Rk is profile core roughness depth [33]. In particular, Rpk may correspond to the highest peaks which will penetrate in the tire and whose part will be worn off or polished under traffic, Rk to the principal working part of the surface subjected to vehicle load, and Rvk to the capacity of the surface to evacuate or store water [34]. Focusing on dense asphalts, in Table 9, it can be seen that for the modified mixes (Dense 50/70 GP and Dense HD) there is an average increase for Rpk and Rk parameters of around 20% and 65%, respectively, if compared with the dense mixture of traditional asphalt (Dense 50/70). About the Rvk parameter, only for the Dense 50/70 GP sample did the value double when compared to Dense 50/70.

For porous mixtures, only the difference between the Porous 35/50 GP mixture and the reference mixture (Porous HD) is relevant: Rpk, Rk and Rvk are decreased by 26%, 10% and 10%, respectively.

As expected, the spectral analysis of the texture shows the substantial

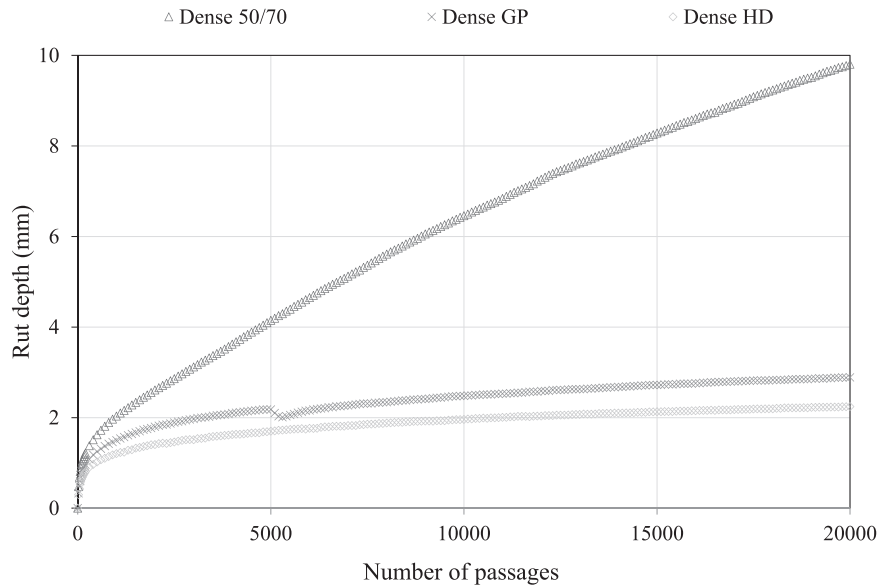


Fig. 9. Trend of rut depth in the wheel-tracking test for dense surface course mixtures.

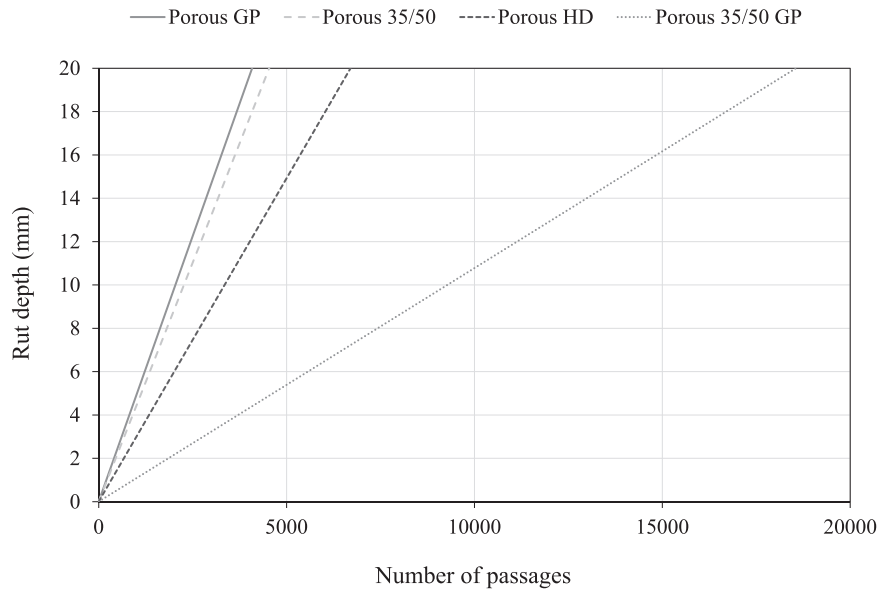


Fig. 10. Trend of rut depth in the wheel-tracking test for porous surface course mixtures.

difference between dense and porous mixtures, see Fig. 13. Instead, with reference to the set of spectra relating to the dense mixtures, there are no elements of great difference between the analysed mixtures. The same consideration can be stated for porous mixtures.

Finally, Table 10 shows the values of friction by British Pendulum and Drainability. All measured values satisfied the technical requirements reported in, namely PTV > 55 and Drain > 18 L/min.

Accordingly, the additive should be described as maintaining compliance with the investigated surface-performance requirements, rather than as having no effect on surface properties. In fact, some indicators, such as PTV for the dense GP mixture, decreased compared with the corresponding reference mixture although they remained above the minimum requirement.

3.4. Life-cycle assessment – “cradle to gate”

As anticipated, also the environmental impacts of the mixtures were

studied by means of LCA approach, with a focus on the production stage. The impacts related to raw materials transport and production and to asphalt mixtures production were calculated for all the mixtures studied, based on primary and secondary data.

The main assumptions adopted for the LCA are summarized in Table 11 and are consistent with previous studies by the authors [24, 25]. In detail, the LCA assumptions adopted in this study are coherent with those already discussed in previous works by the authors [24,25], to be considered for further details on plant productivity, energy consumptions and transport modelling. The reference unit is represented by the declared unit (DU), consisting of 1 metric ton of HMA suitable for surface courses, as described in previous sections. The analysis was carried out according to a cradle-to-gate approach, including raw-material production, transport to plant and asphalt mixture production. The inventory combined mix-specific quantities with plant and logistics data representative of the local context. Accordingly, the environmental results reported here refer to the production stage only

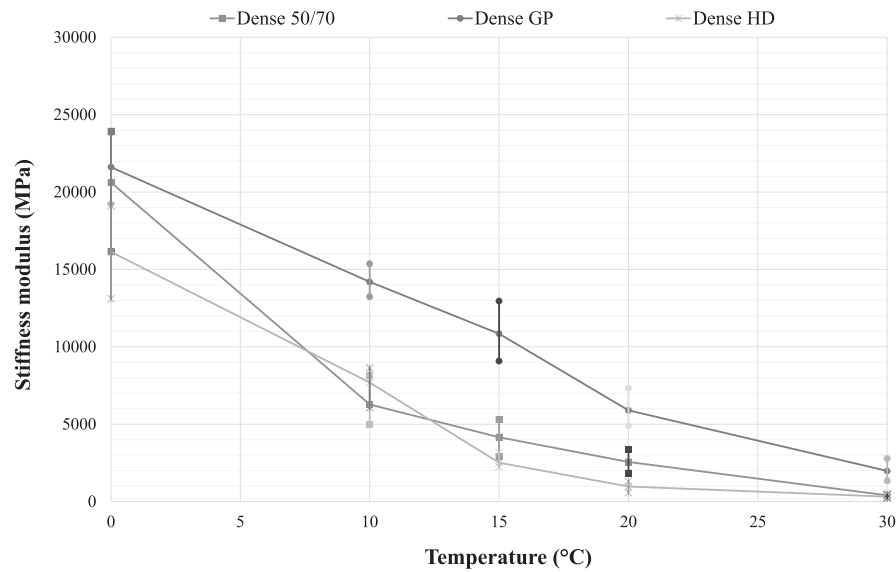


Fig. 11. Stiffness modulus values for dense surface course mixtures.

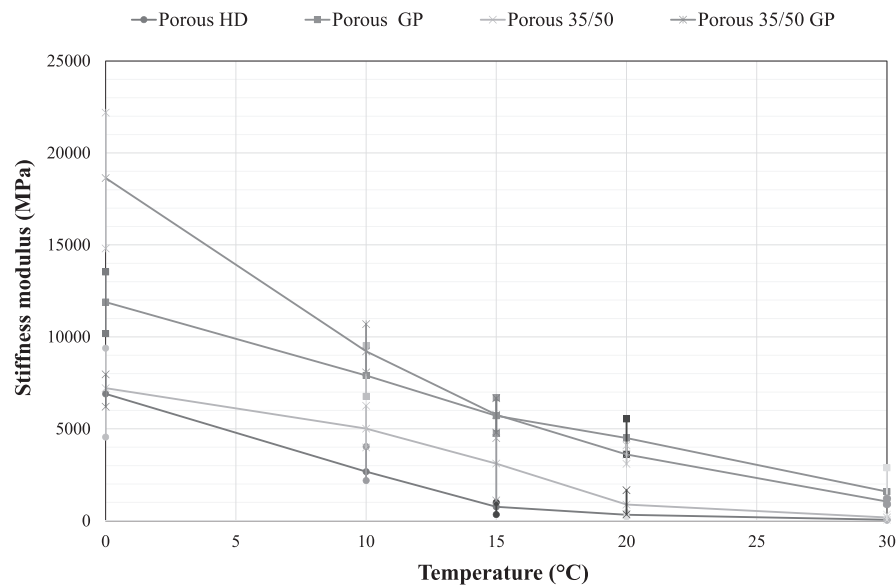


Fig. 12. Stiffness modulus values for porous surface course mixtures.

Table 8

Classical roughness parameters of the mixtures surface analysed.

Parameters	Unit	Dense 50/70	Dense GP	Dense HD	Porous HD	Porous GP	Porous 35/50	Porous 35/50 GP	Standards or Reference
MPDiso	mm	0.951	0.760	0.792	3.135	2.980	3.220	2.671	ISO 13473-1 1997
MPDaipcr	mm	1.029	0.695	0.745	3.037	2.817	3.344	2.791	[25]
ETD1995iso	mm	0.961	0.808	0.833	2.708	2.584	2.776	2.337	ISO 13473-1 1997
ETD2019iso	mm	1.046	0.836	0.871	3.449	3.278	3.542	2.939	ISO 13473-1 2019
ETD1995aipcr	mm	1.023	0.756	0.796	2.630	2.453	2.875	2.433	ISO 13473-1 1997
ETD2019aipcr	mm	1.204	0.816	0.868	3.083	3.109	3.555	3.050	ISO 13473-1 2019
Ra	mm	0.569	0.493	0.507	2.085	1.964	2.175	1.885	ISO 21920-3: 2021
Ru	mm	1.039	0.903	0.882	3.392	3.206	3.425	2.857	
Rz	mm	2.243	1.906	2.124	7.536	7.209	7.925	7.095	
Rmax	mm	3.188	3.182	3.249	11.465	10.539	11.229	10.538	
RMS	mm	0.748	0.694	0.681	2.686	2.495	2.813	2.406	

and allow a consistent comparison among the investigated mixtures. The environmental categories assessed are listed in Table 12.

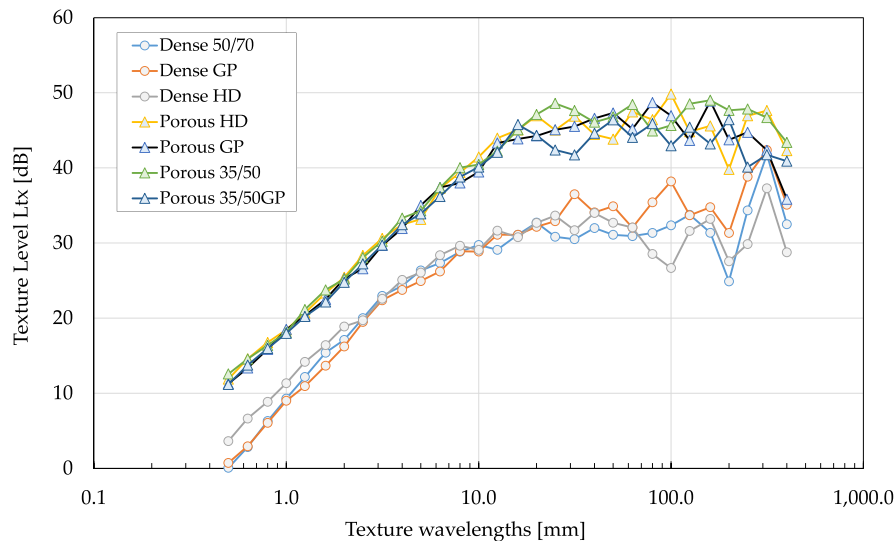
The results indicate that the incorporation of the innovative additive

reduces the production-stage environmental impact by approximately 9% for the dense mixtures and by approximately 10% for the porous mixtures, within the adopted cradle-to-gate boundary. Figs. 14–17 show

Table 9

Statistical parameters of the mixtures surface analysed.

Parameter	Unit	Dense 50/70	Dense GP	Dense HD	Porous HD	Porous GP	Porous 35/50	Porous 35/50 GP	Standards or Reference
Kurtosis Rku	-	1.175	3.434	2.039	1.098	1.221	0.654	1.374	[28,31,32],
Skewness Rsk	-	-1.018	-1.571	-1.091	-0.993	-0.979	-0.937	-1.105	
from Bearing Abbot Curve	g-factor	%	73.500	76.531	75.969	72.781	71.823	71.927	ISO 10844:2021
	Rpk-10%	mm	0.391	0.313	0.315	1.009	0.970	0.916	[33–35]
	Rk	mm	1.059	0.791	0.830	3.744	3.822	3.997	
	Rvk-80%	mm	1.851	2.252	1.814	6.162	5.860	5.592	

**Fig. 13.** Spectral analysis of the texture for selected mixtures.**Table 10**

Surface characteristics values of all mixtures studied.

Characteristics	Unit	Dense	Dense GP	Dense HD	Porous HD	Porous GP	Porous 35/50	Porous 35/50 GP	Standard
Sand height	mm	1.14	1.17	1.18	3.0	2.8	3.2	3.0	UNI EN 13036–1:2010
Macro-texture	High	High	High	High	Very high	Very high	Very high	Very high	
British Pendulum Number PTV		72	61.5	65.3	62.2	71	72	77	UNI EN 13036–4:2011
Drainability	L/min	-	-	-	34.6	30.2	43.7	28.8	UNI EN 13036–3:2006

Table 11

Main assumptions adopted for the cradle-to-gate LCA.

Item	Assumed value / criterion
Plant productivity	70 t/h
LSF oil consumption for aggregate heating	12 kg/DU
LPG consumption for binder heating	0.63 kg/DU
Diesel consumption for loader operations	0.19 kg/DU
Transport distance - limestone aggregate	0.3 km
Transport distance - neat bitumen and diesel	11.7 km
Transport distance - PMB	44.4 km
Transport distance - additive / polymeric compound	1423.0 km
Transport distance - LSF oil	30.0 km

that the innovative mixtures have a lower production-stage environmental impact than the corresponding traditional mixtures under the adopted assumptions. In particular, Fig. 14 and Fig. 15 indicate reductions in the basic environmental impact indicators for traditional mixtures, both with and without additives. A similar trend can be seen in Fig. 16 and Fig. 17, referring to the porous asphalt mixtures with and without additives.

The graphical representation indicates that the reduction in production-stage environmental impact is not confined to a single

Table 12

Environmental categories investigated.

Categories		Unit
Acidification	AC	[kg SO ₂ eq]
Eutrophication	EU	[kg PO ₄ eq]
Global warming	GW	[kg CO ₂ eq]
Photochemical oxidation	PO	[kg NMVOC]
Abiotic depletion, elements	AD	[kg Sb eq]
Abiotic depletion, fossil fuels	AD*	[MJ]
Water scarcity	WS	[m ³ eq]
Ozone layer depletion	OD	[kg CFC ⁻¹¹ eq]
Carbon monoxide	CO	[g]
Lead	Pb	[mg]
Mercury	Hg	[μg]
Nitrogen oxide	NO _x	[g]
Particulate matter	PM10	[g]

category but extends across multiple indicators. This suggests that the observed benefit is mainly linked to the modification of material composition, rather than to isolated variations in the production process. These reductions can be achieved by partially replacing virgin bitumen with an engineered additive containing graphene and recycled

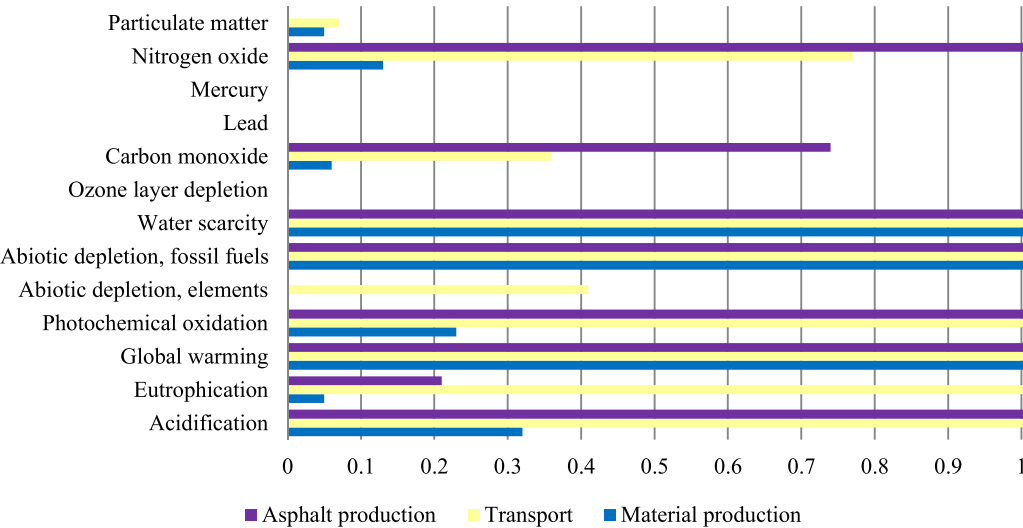


Fig. 14. Environmental impacts for dense mix.

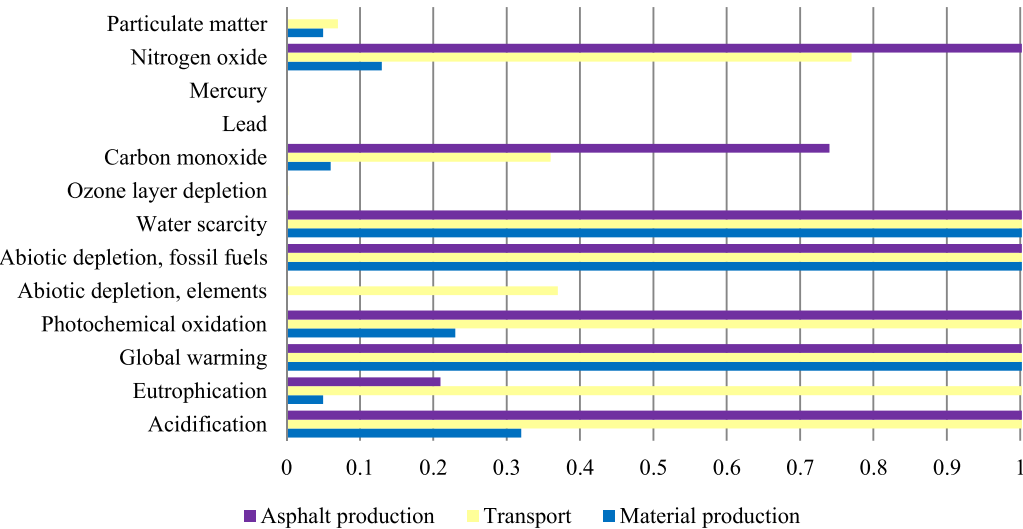


Fig. 15. Environmental impacts for dense mix with graphene-based additive.

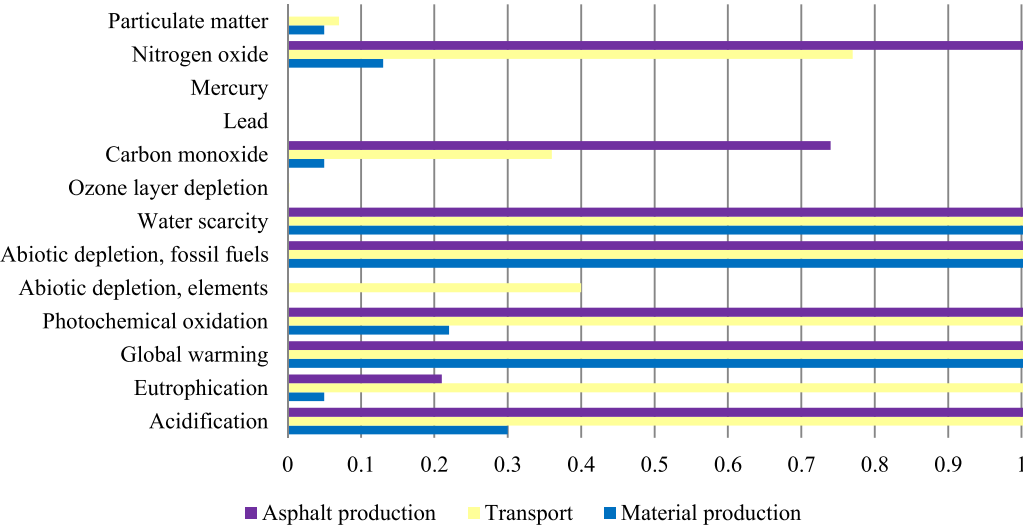


Fig. 16. Environmental impacts for porous mix.

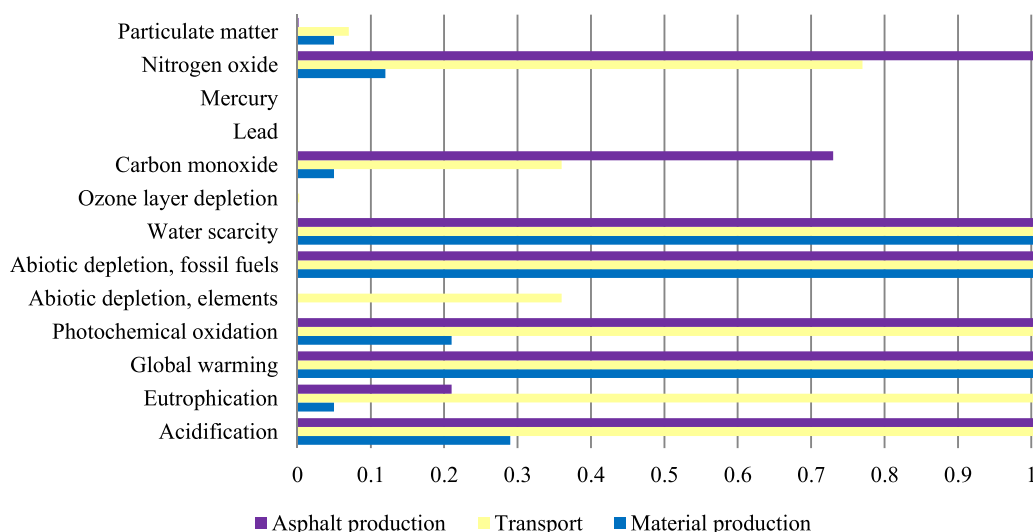


Fig. 17. Environmental impacts for porous mix with graphene-based additive.

polymer fractions. As bitumen production contributes significantly to both global warming potential and the depletion of fossil resources in asphalt mixtures, reducing demand for binders, even by a small amount, can produce measurable environmental benefits.

Therefore, achieving a 9–10% reduction without altering the technology used in the plants or introducing other upstream processes is a notable result within the declared-unit production-stage assessment. However, material optimisation alone cannot be used to infer whole-life environmental superiority without additional service-life and maintenance data.

The environmental improvement observed in this study is related to reduced energy consumption rather than changes in production factors. This supports the interpretation that the environmental benefit mainly comes from the different composition of the materials and the reduced need for virgin binders within the cradle-to-gate boundary.

From the environmental viewpoint, the partial replacement of virgin bitumen with graphene-reinforced recycled polymeric material produced a measurable cradle-to-gate benefit.

The improved laboratory performance suggests that service-life extension may be a possible hypothesis, but this inference requires confirmation through fatigue testing, moisture susceptibility assessment, low-temperature cracking tests, ageing assessment, field monitoring, and whole-life analysis.

The results support the integration of performance-oriented material innovation and circular economy principles into asphalt pavement engineering, provided that long-term durability and whole-life environmental performance are confirmed by further studies.

Future research should therefore investigate long-term durability, moisture susceptibility, low-temperature behaviour, fatigue performance, and field constructability, while extending the assessment to whole-life environmental and economic dimensions.

4. Conclusions

This study aimed to evaluate the impact of adding a graphene-based additive containing recycled polymers on the volumetric, mechanical, functional and environmental performance of asphalt wearing-course mixtures.

The gyratory compaction tests confirmed that the modified mixtures exhibit acceptable workability and densification behaviour. The investigated surface-related properties, such as skid resistance, macrotexture and drainability, remained within the expected technical range for the analysed mixture families, although some indicators varied with respect to the reference mixtures. The mechanical performance of the mixtures

investigated showed improvements in terms of ITS and ITSM, with increases of about 50% for the selected mechanical indicators. Furthermore, tests conducted for permanent deformation highlighted enhanced resistance, with substantial improvements in performance also observed in porous mixtures.

From the environmental perspective, measurable cradle-to-gate benefit can be ensured by the partial replacement of virgin bitumen with graphene-reinforced recycled polymeric material. It should be noted that the environmental analysis was limited to the production phase; however, the LCA study demonstrated a reduction in the environmental impact of traditional mixtures of approximately 9% and porous mixtures of approximately 10%.

The structural properties were enhanced in the investigated laboratory tests, which suggests that service-life extension may be a possible hypothesis; however, this inference still requires confirmation through fatigue testing, moisture susceptibility assessment, low-temperature cracking tests, ageing assessment, field monitoring, and whole-life analysis. Only after such validation could possible additional sustainability benefits be assessed within a whole-life framework. The results support the integration of performance-oriented material innovation and circular economy principles into asphalt pavement engineering, provided that long-term durability and whole-life environmental performance are confirmed by further studies.

Future research should further investigate long-term durability, moisture susceptibility, low-temperature behaviour, fatigue performance, and field constructability. Furthermore, the optimum dosage of the graphene-based additive will be investigated, for improved applications. Finally, future investigations will aim to extend the assessment to whole-life environmental and economic dimensions, to include construction, use, maintenance, and end-of-life phases and, thus, provide a more comprehensive evaluation of environmental benefits.

Data availability statement

Some or all data, models, or code generated or used during the study are proprietary or confidential in nature and may only be provided with restrictions.

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Declaration of Competing Interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Clara Celauro reports supplies (i.e. the additive) was provided by Iterchimica srl. The other 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.

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Data availability

The data that has been used is confidential.

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