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Investigating the environmental and mechanical properties of sustainable asphalt mastic solutions for road flexible pavements

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Abstract

The circular economy is restorative in nature, and it aims to maintain the utility of products, components and materials for as long as possible while also retaining their value. It thus minimizes the need for new inputs of virgin materials and energy, while reducing environmental pressures linked to resource extraction, emissions and waste management. The research focus on the investigation of an innovative procedure for reusing the jet grouting waste (JGW) obtained from soil consolidation activities to produce high-performing sustainable road asphalt mastics (filler plus neat bitumen). The mastics under analysis were made at three different filler over bitumen ratios of 0.5, 1 and 1.5, in particular six were obtained with neat bitumen and other six at the same f/b ratios mixed with an aged bitumen after the RTFO procedure. The rheological properties of the mastics were investigated using a dynamic shear rheometer, and the performance of the innovative mastics was compared with that of the neat bitumen and traditional mastics made up of bitumen and limestone filler (LF). The main results of the initial step of the analysis showed that, at temperatures from 0 to 30°C, the shear moduli of the mastics containing JGW (MJGW) averaged 30% higher than the limestone mastic (ML) blended with an f/b ratio of 0.30. The life-cycle assessment impact category indicators were then calculated, highlighting the considerable environmental benefits of the innovative hot asphalt mastics in question. The primary outcomes showed that compared with the asphalt mastics containing LF, the asphalt mastic solution lowered terrestrial ecotoxicity by 53% and the freshwater eutrophication potential by up to 97%.

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Introduction

Bituminous binder and filler, consisting in finer powder of an aggregate gradation for road asphalt concrete mixtures and passing from 70% to 100% at 0.063mm sieve (EN 13043), constitute one mixed so-called mastic (binding phase) that really influences the properties of hot-mix asphalt (HMA) such as workability, moisture sensitivity, stiffness, resistance to permanent deformation at high temperatures, fatigue life at intermediate temperatures and cracking resistance at low temperatures. HMA should be considered consequently a mixture comprising mastic-coated aggregates rather than pure bitumen-coated aggregates (Buttlar et al., 1991).

Filler is used in HMA for contributing to increase rutting resistance deformation, fatigue cracks and moisture failures, also decreasing the air void content (Viscione et al., 2022). As a result, bituminous mastic contributes to larger inner cohesion of a mixture as a whole as well filler type and its relative content affect it: appropriate filler over bitumen content will help to reach good final mechanical properties of the asphalt mixtures since large amount of filler can cause not very workable final mixture while its insufficient amount can negatively affect the stability conditions of the final mixture.

Fillers vary in physical and chemical properties, shape, texture, size and gradation; thus filler choice is very crucial action for ensuring keeping ideal performance of HMA (Veropalumbo et al., 2019).

Some scientists investigated for example (Faheem et al., 2010) the relationships between filler's chemical composition and mastic behaviour founding that the chemical composition of filler does not contribute directly to the filler stiffening effect in mastic; the maximum amount of filler that can be mixed with the bitumen is quasi-single dependent on the particle geometry characteristics of the filler, which are synthetized by the Rigden voids value, and there is little influence from the bitumen.

Nowadays due to environmental and economic concerns, the use of recycled waste materials in road pavements has considerably extended to decrease the need of using natural resources and hence to prevent some irreparable environmental destruction also by decreasing the impact of waste storage problems on the environmental landscape.

Antunes et al. (2017) investigated the adoption of three different waste materials (construction and demolition waste, brick powder and fly ash) as filler in substitution of limestone one in asphalt mixtures. The absorption test and the delta ring and ball test were carried out to investigate the interaction between the waste filler and bitumen through the mastics. The results demonstrated that the mastics with waste materials showed strong resistance to water damage in particular when the fly ashes are used in substitution of limestone filler since it results in a strong bitumen bonding due to the singular characteristics of its particles. Also, Choudhary et al. (2021) demonstrated that the moisture resistance, bonding and adhesion in asphalt mixes are greatly influenced by the mineralogical composition of its filler particularly when filler is a calcium-based minerals.

To reduce the pollution problem Meng et al. (2022) investigated through a rheological perspective the possibility of using steel slag powder (SSP) and crumb rubber, that are wastes brought by industrial development, into asphalt to make asphalt mastic. Rheological tests performed by means of a dynamic shear rheometer showed that the SSP as filler enhanced the performance of asphalt mastic at higher temperatures and reduced temperature sensitivity than a traditional limestone mastic.

The adoption of waste fillers into asphalt mastics do not produce only mechanical benefits but also an environmental advantage. The useful tool to assess the environmental issues concerning the waste materials into asphalt pavements is the Life cycle assessment (LCA). Many researchers are focusing the attention on the LCA. An example is Yao et al. (2022) that by performing an LCA found that replacing the SBS modified asphalt with PET modified asphalt in asphalt pavement could save about 7.3% of the greenhouse gas emission (GHG) emissions.

These values are about 9% and 4% higher than the control values for the two indicators, respectively. The study recommended 40% partial replacement of granite with RCA in Stone Mastic Asphalt for optimum strength and environmental impact.

Aghayan et al. (2021), instead, showed that by using ceramic waste materials and coal waste powder in substitution of traditional limestone aggregates reduced about of 10% the greenhouses. Also, the combination of these two wastes showed the highest positive impact on the global warming index, reducing by 9 %.

The research presented here aims to explore two main questions considering a) the environmental feasibility of an alternative filler for making asphalt mastics, today classified as a waste and going to landfills unless otherwise stated in the technical specifications, and b) the evaluation of the optimum filler over bitumen ratio to suggest in asphalt

concrete blends by varying filler type and downstream the assessment of rheological properties of the mastics and binders. The jet grouting waste (JGW) as filler deriving from the consolidation activities of the underground walls was investigated as substitution of traditional limestone filler (LF) for asphalt pavements by analyzing the rheological properties of asphalt mastics at three different filler-over-bitumen ratio (f/b) of 0.5, 1 and 1.5. Aging effect in a short-term of the bitumen here used was examined to determine, first of all, if the hardening as a consequence of the aging can be considered slight and not significant affecting the rheological response of the mastics otherwise it is desirable to find what filler type, and consequently a proper filler over bitumen ratio, that can reduce the effect of aged bitumen that's actually caused at plant during a blend mixing phase.

The rheological performance were assessed through a dynamic shear rheometer by evaluating the stiffness as result of the frequency sweep test and the resistance to permanent deformation as outcomes of a multiple stress creep and recovery test. The environmental compatibility of the designed asphalt mastics when used as bonding structure of an asphalt mixtures was analysed through the LCA.

2. Materials and methods

2.1. Fillers and bitumen

In this research, two different fillers were selected for this investigation: limestone filler (LF), and jet grouting waste (JGW). The limestone filler is the traditional material employed in the production of bituminous mixtures for roads superstructures and derives from limestone crashing, made into fine powder by a suitable sieving in order to use it as a filler. The jet grouting is the jet grouting waste used for the consolidation of the walls of tunnels after that this have been excavated. Those materials have been classified as filler since the requirements were found to be compliant to the % values specified by EN 13043.

The Scanning Electron Microscope (SEM) that have the ability to work at high magnifications by using a beam of electrons, was used to investigate the sample through very small details, in particular micron size (μm). Figure 1 returns the observation of the morphology of the particles for the examined fillers; in particular, Table 1 summarizes their form and size.

Table 1 Morphological description of the fillers.

Filler	Morphology	Size (diameter)
Limestone	Particles at contours jagged both smooth and wrinkled with spherical elements, also aggregated each other, with the presence of rare pores*	10 ÷ 76 mm (jagged elements) 3 ÷ 55 mm (spherical elements)
Jet Grouting	Particles with smooth and wrinkled sharp edges	2.7 ÷ 56.1 mm

* the presence of fine particles, even aggregated, can also be observed, whose size varies from nanometric levels up to 10 mm

A neat bitumen 50/70 penetration grade (B5070) was used in this study to prepare asphalt mastics. Table 2 shows the main properties of the binder that are in agreement with European Standards.

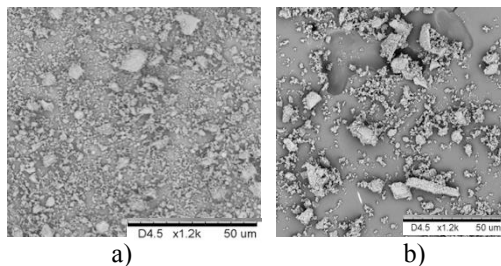


Figure 1 SEM images. a) LF and b) JGW.

Table 2 Binder properties.

Test	Unit	Value	Standard
Penetration @ 25 °C	dmm	61	EN 1426
Soft. point	°C	48.8	EN 1427
Dynamic Viscosity @ 160 °C	Pa s	0.22	EN 13702
Frass	°C	-10	EN 12593

Asphalt binders age primarily due to two different mechanisms: volatilization of light oils present in asphalt and oxidation by reacting with the oxygen in the environment. The blending and agitation in the hot mixing facility and during placement ages the binder by both mechanisms because of the high temperature and air flow involved in the

process. To simulate this form of aging in compliance with the SuperPave specification, in the present research, to simulate the aging effect the Rolling Thin Film oven procedure (EN 12607-1) was carried out.

2.2. Sample preparation

Six mastics were prepared by blending each study filler (LF and JGW) with the bitumen using the filler over bitumen ratio hypothesized of 0.5, 1 and 1.5 by adopting both original bitumen (B5070) and aged-RTFO bitumen (AB5070) achieving six blends for each filler type as follows:

- LF plus B5070 at f/b equal to 0.5, 1 and 1.5 to obtain respectively LM0.5, LM1 and LM1.5
- LF plus AB5070 at f/b equal to 0.5, 1 and 1.5 to obtain respectively ALM0.5, ALM1 and ALM1.5
- JGW plus B5070 at f/b equal to 0.5, 1 and 1.5 to obtain respectively JGM0.5, JGM1 and JGM1.5
- JGW plus AB5070 at f/b equal to 0.5, 1 and 1.5 to obtain respectively AJGM0.5, AJGM1 and AJGM1.5

A RW 20 DZM.N mechanical mixer module (IKAWerkeGmbH & Co. KG, Staufen, Germany) was used to blend the filler and asphalt binder at mixing temperatures of 160°C by following previous mixing procedures (Veropalumbo et al., 2020).

2.3. Methods

2.3.1. Rheological investigation

Laboratory investigations based on rheological measurements showed that mastics can be considered thermorheologically simple linear viscoelastic materials and the mechanical influence of fillers can be assessed through a viscoelastic analysis of the mastics.

To evaluate the viscoelastic properties of the binders, in particular of the mastics, in a determined range of temperatures and frequencies the DSR equipment was used which operates in the oscillatory system, where the load does not remain constant over time but varies according to a predefined law, usually of the sinusoidal type, which better simulates traffic effects.

The procedure with a test in oscillatory shear involves determining the complex shear modulus (G^*) and phase angle (δ) of binders over a range of test frequencies and test temperature (EN 14770 standard). For each of the nine mastics packaged, the values of the G^* were investigated at five different temperatures between 10 and 50 °C with increments of 10 °C, and 19 different frequencies (from 0.1 to 10 Hz). The value of the shear stress imposed carrying out the test was defined in compliance with the Annex C of EN 14770. The determination of G^* and δ was obtained in parallel plate configuration with 8mm diameter and a gap 2 mm gap for the temperature lower than 30 °C instead for the temperature higher than 30 °C was adopted a 25mm diameter with 1 mm gap.

To determine the presence of elastic response in bitumen and bituminous binders the multiple stress creep and recovery (MSCR) was performed under two stress levels of 0.1 and 3.2 kPa at the temperatures of 40 and 60 °C. The presence of this elastic response is determined by measuring the non-recoverable compliance (J_{nr}) of the binder and mastics. The test consists of to load the sample at constant stress for 1 s, then allowed to recover for 9 s. Ten creep and recovery cycles were run at 0.100 kPa creep stress followed by 10 more cycles at 3,200 kPa creep stress.

2.3.2. Life cycle assessment

LCA addresses the environmental aspects and potential environmental impacts (e.g. use of resources and the environmental consequences of releases) throughout a product's life cycle from raw material acquisition through production, use, end-of-life treatment, recycling and final disposal; LCA can be applied to make environmental claims regarding the superiority or equivalence of one product versus a competing product that performs the same function (EN 14040).

LCA was applied to a local case study where the asphalt mastics here designed were used as the bonding structure of an aggregate skeleton making up the asphalt mixture used to rehabilitate, repair or rebuilt a road asphalt pavement. As suggested by the standardization framework of EN 14040, LCA was performed according to 4 main steps: the goal

and scope definition, the life cycle inventory analysis, the life cycle impact assessment and the final interpretation of the results.

The system boundary was defined according to the guidelines of EN 15804, including the production and supply of natural resources (aggregates, fillers, secondary raw materials (JGW), bitumen), energy, the asphalt mixture production in a centralized asphalt plant powered by natural gas and electricity, the transportation of the asphalt mixture to the construction site, the laying and compaction of the asphalt layer, the demolition and disposal of the asphalt layer at the end of the service life.

The life cycle inventory was drawn using surveyed primary data (amount of materials needed to manufacture each asphalt mixture, machinery's productivity and diesel fuel consumption, electricity and natural gas consumption of the asphalt plant) and secondary data (from Ecoinvent 3.8 database (Wernet et al., 2016) and Eurobitume association (Blomberg et al., 2011)), while impact assessment was performed using SimaPro 9® calculation tool. The selected input and output flows of materials and energy were converted into 18 environmental impact category indicators according to ReCiPe 2016 Hierarchist (H) method (Oreto et al., 2021) that converts input and output flows into midpoint damage categories placed midway along the damage path.

3.1 Mechanical behaviour

In Table 3 are shown the maximum and minimum G^* shear modulus values, as results of the frequency sweep test. The maximum achieved values of G^* at each f/b were showed, for AJGM1 and AJGM1.5 (130, and 243.3 MPa) instead the minimum values at each f/b were returned by JGM0.5, LM1 and LM1.5 (0.003682, 0.005734 and 0.01685 MPa).

Such results indicate that the complex shear modulus of asphalt binder is improved when the mineral fillers are used. The increase in the G^* for mastics with high filler content is caused by fillers contributing such a large volume to the binder that they become the predominant component in bitumen–filler mastics. The effect of f/b ratio on the complex shear modulus was significant, i.e., the G^* increases exponentially as the f/b ratio increases. As showed in Table 3 the variability of the results in terms of G^* are less spread out for $f/b=1.5$ for the mastics containing JGW when aged, in fact the lowest CV value of 1.62 was found for the AJGM1.5 solution.

Table 3 Summarize of the G^* values from frequency sweep test performed on the asphalt mastics.

	f/b											
	0.5				1				1.5			
	Max	Min	CV	Dev.st.	Max	Min	CV	Dev.st.	Max	Min	CV	Dev.st.
G^* _LM	48.46	0.004206	1.89	9.52	66.88	0.0057	1.86	13.16	173.50	0.0168	1.89	35.29
G^* _JGM	52.79	0.00602	1.73	11.02	121.3	0.0117	1.76	25.03	160.30	0.0439	1.48	38.67
G^* _ALM	53.88	0.005534	1.74	11.05	93.95	0.0125	1.64	20.89	204.00	0.0267	1.63	47.34
G^* _AJGM	60.97	0.003682	1.86	11.96	130	0.0073	1.82	26.84	243.30	0.0170	1.62	53.82

The combination alternative filler-bitumen resulted an increasing of G^* compared to mastics containing limestone filler. In fact, JGM for the temperature higher than 30 °C exhibits an increase in terms of G^* of 0.70%, 14.74% and 0.52% respectively at $f/b=0.5$, 1 and 1.5 compared to LM. As regards the test temperatures lower than 30 °C, JGM shows an increase of G^* respectively of 37.31%, 53.07% and 53.93% at $f/b=0.5$, 1 and 1.5 respect to LF (see Figure 2). On the other hand, as the filler concentration increases, the influence of the filler type becomes more apparent, which implies that the influence of the filler on the asphalt binder is dependent on the filler type.

The aging of the bitumen produced mastics with high G^* values. In fact, the G^* values of ALM and AJGM at f/b equal to 0.5, are respectively 11.18 and 15.5 higher than that of LM and JGM, while at f/b equal to 1 are respectively 40 and 7% higher, and 17.6 and 51.7% higher at f/b equal to 1.

Substantially, among all mastics solutions under exam, AJGM1.5 showed the highest G^* values for test temperatures above 30 °C.

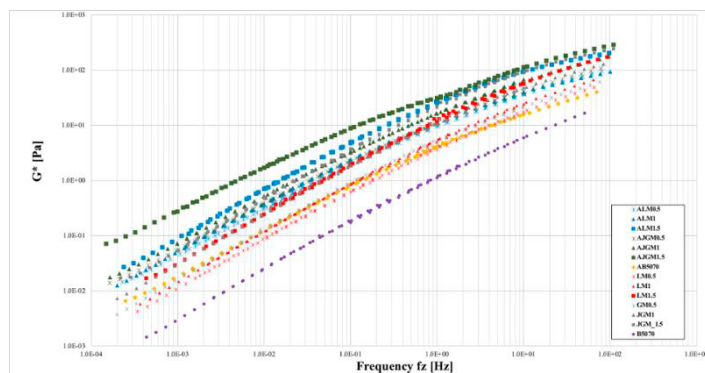


Figure 2 Master curves results.

A dynamic shear rheometer (DSR) was used to evaluate the rutting potential of laboratory produced bituminous mastics at high temperatures. Rutting behaviour was evaluated through multiple stress creep recovery (MSCR) tests according to EN 16659.

In order to predict the rutting behaviour of the mastics, the J_{nr} values were analysed. In Figure 3 are reported the J_{nr} values obtained from the investigation of the mastics containing LM and JGM at both test temperatures of 40 and 50 °C. At the temperature of 40 °C, the mastics at $f/b=1.5$ were stiffer, and therefore it was not possible to measure to perform the MSCR.

The obtained results proven that the J_{nr} value for each f/b ratio increases as the temperature increases. In fact, for the test temperature of 60 °C it was showed a J_{nr} reduction of about 98% in comparison to the values obtained at 40 °C regardless the f/b ratio. This result highlights the viscous behaviour of the mastics as the temperature increase. In particular, at 60 °C all the mastics packaged at $f/b=1$ show a decrease of about 69% in comparison to $f/b=0.5$, while the mastics packaged at $f/b=1.5$ exhibited a mean decrease of 90% than $f/b=1$.

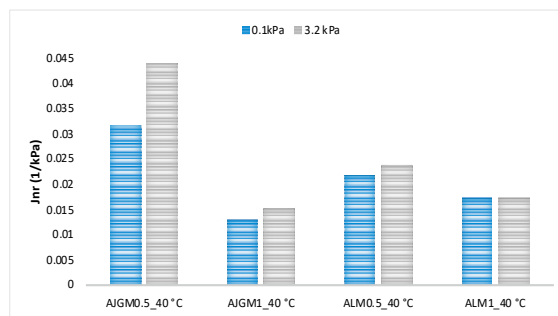
Clear is the reduction of J_{nr} value for the mastics containing limestone filler, that increase as the f/b ratio increases. As shown in Figure 3, JTM decreases of 4%, 3.7% and 46.7% respectively for $f/b=0.5$, $f/b=1$ and $f/b=1.5$ compared to LM at the same f/b ratio.

These results demonstrate the effect of the filler in a composition filler-bitumen where the increases of the added filler in the mastic reduced the non-recoverable creep compliance.

The rutting resistance analysed on the aged mastics confirms that the J_{nr} value increases with the increases the temperature and decreases with the increases of the ratio f/b . Also, for the aged phase, the mastics package with JGWM show a lower J_{nr} value respect to the other, followed by limestone filler. The stiffening effect has been confirmed for the mastics containing alternative materials in the ratio $f/b=1.5$ with a lower J_{nr} ratio, followed by $f/b=1$ and $f/b=0.5$. In finally the capacity of recoverable the deformation, at $F/A=1.5$ showing a capacity to recoverable higher than the other two mastics investigated, both at 40° and 60°C (see Figure 3).



a)



b)

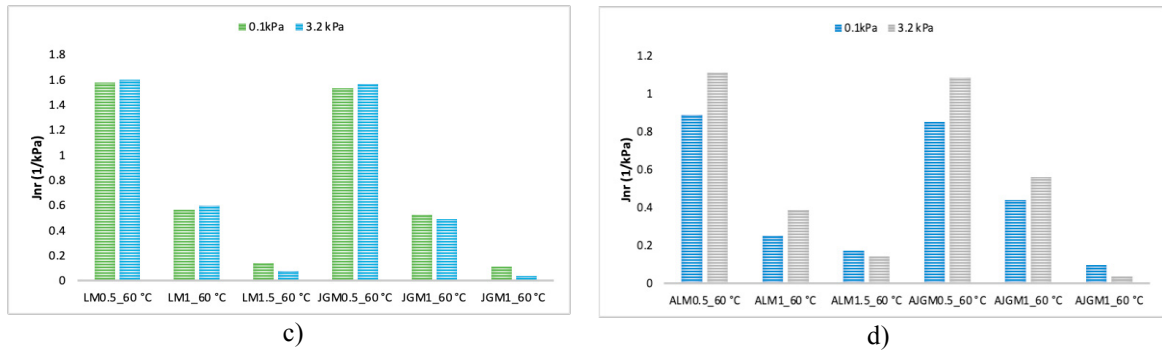


Figure 3 MSC results: a) Unaged mastics at 40 °C, b) Aged mastics at 40 °C, c) Unaged mastics at 60 °C and d) Aged mastics at 60 °C.

3.2 LCA results

The LCA analysis was performed in the case of rehabilitating, repairing or rebuilt of a road pavement where the asphalt mastics at $f/b=1.5$ were considered as bonding structure of the asphalt mixture. The description of the system boundaries and the Life cycle inventory followed previous studies (Oreto et al., 2021).

The overall results for the midpoint indicators of the comparative LCA assessment between the traditional solution with LF and the alternative one made up of JGW are shown in Table 4. The results showed that JGWM has lower environmental impact indicators (averaging - 45.6%) than the traditional LM.

In particular, the JGWM achieves an average reduction of 63.9% for all the impact category indicators than LM; specifically, it can be observed a huge reduction of 49% in terms of a) Terrestrial ecotoxicity, mainly due to gaseous emissions such as NO_x arising from transportation, b) water, which is 84% lower in the JGW recycling process compared to LF production, and c) Freshwater ecotoxicity, which is 97% lower as a result of the smaller amount of phosphorous compounds released in water during JGW crushing compared to LF production.

Table 4 LCA results.

Impact category	Units	Asphalt solutions with	
		LM	JGWM
Global warming	kg CO ₂ eq	9.72E+00	1.69E+00
Stratospheric ozone depletion	kg CFC11 eq	1.03E-05	1.13E-06
Ionizing radiation	kBq Co-60 eq	3.28E+00	1.71E-01
Ozone formation, Human health	kg NO _x eq	4.98E-02	9.61E-03
Fine particulate matter formation	kg PM _{2.5} eq	2.13E-02	2.50E-03
Ozone formation, Terrestrial ecosystems	kg NO _x eq	5.07E-02	9.79E-03
Terrestrial acidification	kg SO ₂ eq	4.90E-02	5.73E-03
Freshwater eutrophication	kg P eq	3.44E-02	1.20E-04
Marine eutrophication	kg N eq	3.28E-04	1.03E-05
Terrestrial ecotoxicity	kg 1,4-DCB	6.82E+01	2.96E+01
Freshwater ecotoxicity	kg 1,4-DCB	8.28E-01	4.22E-02
Marine ecotoxicity	kg 1,4-DCB	5.29E+03	3.59E+02
Human carcinogenic toxicity	kg 1,4-DCB	1.85E+03	2.53E+00
Human non-carcinogenic toxicity	kg 1,4-DCB	8.33E+03	2.91E+02
Land use	m ² a crop eq	1.50E+00	6.56E-02
Mineral resource scarcity	kg Cu eq	2.89E-02	6.20E-03
Fossil resource scarcity	kg oil eq	3.86E+00	5.94E-01
Water consumption	m ³	3.95E-01	2.59E-03

4. Conclusions

The present study showed through a rheological investigation of mastics and their environmental application how alternative materials, such as jet grouting waste, substituted to ordinary limestone aggregates can be used in the construction and/or maintenance of road superstructures. Six mastics were analysed at three different filler over

bitumen ratios obtained with neat bitumen and other six mastics at the same f/b ratio mixed with an aged bitumen after the RTFO procedure. The LCA analysis was carried out for further comparing alternatives and determining how many sustainable improvements can be derived by substituting the LF with JGW into an ordinary asphalt mixture.

The main results obtained can be summarized as follow:

- The frequency sweep results expressed through the master curves confirms the high stiffness for the alternative mastics at high temperatures and low frequencies of load of application.
- The MSCR show a reduction of the permanent deformation from low temperatures to high temperatures. The Jnr value confirm the stiffening role of the jet grouting waste at the filler over bitumen ratio of 1.5
- The effect of the aging on the bitumen has led an increments of the stiffness in the mastics clear in the comparison between the unaged mastics and aged mastics in terms of shear modulus G^* and permanent deformation.
- LCA results highlighted that JGW recycling is a more environmentally-sound recycling strategy than LF

The results obtained not only confirmed the possibility of replacing the traditional limestone material used in the production of asphalt mixtures, but have shown that with a decrease in the quantity of bitumen and an increase of these materials there is an increase in the mechanical performances usually required by a traditional composition limestone filler-bitumen. These results are the incipit of a subsequent analysis regarding the evaluation of volumetric and mechanical properties of HMA, validating them in comparison to the limits imposed by the most common specifications adopted for the prequalification of a mixture for a road surface.

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