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Analysis of the influence of multi-source waste-based additives on the manufacturing of warm temperature bituminous mixtures

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ABSTRACT: Growing environmental awareness of the depletion of finite natural resources (gas, fuel, etc.) and the continuous rise in greenhouse gas emissions (NO_x, CO, CO_2, etc.) has driven the road engineering industry to develop technologies for producing asphalt mixes at lower temperatures than traditional hot mix asphalt (HMA). Warm mix asphalts (WMAs) offer a viable alternative to conventional mixes, maintaining both mechanical performance and durability while providing a positive ecological impact. This paper presents an investigative study aimed at analyzing the reuse of waste by-products in the production of WMAs at 140°C. To achieve this, three different waste materials (a zeolite by-product from the petrochemical industry, charred acacia splinters, and charred plastic) were characterized and analyzed as additives for the indirect foaming of WMA. Their mechanical performance was then compared to a reference HMA. The results showed that the zeolite by-product exhibited similar workability and compaction to traditional HMA produced at 160°C, while also demonstrating comparable stiffness modulus, resistance to plastic deformation, and water resistance.

KEY WORDS: Asphalt mixtures; additives; Indirect foaming; Waste by-products; Warm mix asphalt.

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RESUMEN: *Análisis de la influencia de aditivos basados en residuos de múltiples fuentes en la fabricación de mezclas bituminosas tibias.* La conciencia ambiental sobre el impacto del agotamiento de recursos naturales finitos y las constantes emisiones contaminantes de gases de efecto invernadero (NO_x, CO, CO_2, etc.) han llevado a la industria de la ingeniería vial a desarrollar tecnologías de mezclas asfálticas producidas a temperaturas reducidas en comparación con el asfalto convencional en caliente (HMA, por sus siglas en inglés). Las mezclas asfálticas tibias (WMAs, por sus siglas en inglés) constituyen una sólida opción para reemplazar las mezclas convencionales, manteniendo tanto el rendimiento mecánico como la durabilidad, mientras se genera un impacto ecológico positivo. Este artículo presenta un estudio investigativo con el objetivo de analizar la reutilización de subproductos de desechos en la producción de WMAs a 140 °C. Para lograr este objetivo, se caracterizaron y analizaron tres materiales de desecho diferentes (subproducto de zeolita de la industria petroquímica, astillas de acacia carbonizadas y plástico carbonizado) para ser utilizados como aditivos para la espumación indirecta de WMAs y, posteriormente, se comparó su rendimiento mecánico con un HMA de referencia. Los resultados concluyeron que el subproducto de zeolita mostró una trabajabilidad y compactación similares al HMA tradicional fabricado a 160°C, conservando su módulo de rigidez, deformación plástica y resistencia al agua.

PALABRAS CLAVE: Mezclas asfálticas; Aditivos; Espumación indirecta; Subproductos de desecho; Mezcla asfáltica tibia.

1. INTRODUCTION

Among industrial activities, construction is connected with a growing greenhouse gas emission, (such as NOx, CO, CO_2, among others) accounting for at least 10% of the total (1) and making up 23% of the overall CO_2 emissions generated by global economic actions (2). Asphalt mixtures are the most common material used for paving roads, contributing to up to 300 million tons of production in 2021 in Europe alone (3). Given the substantial consumption of natural mineral aggregates in the production of bituminous mixtures, comprising nearly 95% of the overall mixture weight, researchers have progressively focused their efforts on exploring the possible replacement of these aggregates with locally available waste materials or by-products, such as recycled crumb rubber (4, 5), slags from different industries (6, 7), construction and demolition waste (8), plastic (9), among others. This approach adheres to the principles of the circular economy by serving multiple purposes, including reducing dependence on finite natural resources, lightening the buildup of waste in landfill sites, and promoting essential economical industrial symbiosis (10).

Regular hot mix asphalt (HMA) manufacturing conditions can reach up to a mix temperature of around 165°C which makes it a heavy thermal energy-consuming industry (11). Moreover, the elevated temperatures required for heating the materials during the mixing of bitumen and mineral aggregates contribute to the emission of greenhouse gases and other pollutants that adversely affect air quality, such as sulfur dioxide (SO_2), volatile organic compounds (VOCs), and nitrogen oxides (NO_x), among others. For this reason, and to incorporate sustainability criteria, it is essential to seek a decrease in the temperatures at which asphalt pavements are manufactured. Henceforth, asphalt mixes produced at warm temperatures (WMA) constitute an interesting alternative to be considered allowing a decrease in manufacturing temperature by approximately 20–40°C when compared to typical HMA, this reduction mitigates its adverse ecological effects without potentially hampering its overall performance (12, 13). The environmental benefits of Warm Mix Asphalt (WMA) production have been widely discussed in the literature, particularly regarding its potential to reduce emissions during production. For example, studies (13, 14) report that reductions in CO_2 emissions can range from 15% to 46%, while reductions in SO_2 emissions vary between 20% and 35%, along with reductions in other pollutants. These variations underscore that the extent of emission reductions depends on multiple factors, such as the specific production methods, additives used, and operational conditions. This variability highlights the complexity of quantifying the environmental benefits of WMA without a detailed understanding of the specific processes involved. Furthermore, the reduction in binder temperature contributes to less volatiles being lost and diminished bitumen oxidation, both of which are linked to material aging. Additionally, maintaining the inherent characteristics of bitumen throughout the production process results in a more glutinous material that exhibits reduced vulnerability to fragile failure (15).

WMA can be categorized into three groups based on the applied methodology to reach a lower required mixing temperature: first, by the inclusion of bio-based additives (Fischer-Tropsch wax, Montan Wax, etc.), or chemical additives (emulsification agents, surfactants, etc.), and the introduction of water to induce asphalt foaming (13). Organic additives, typically waxes and fatty amides, function by lowering the viscosity of the binder when it exceeds its melting point. In contrast, chemical additives, which are generally emulsifiers and surfactants, do not affect binder viscosity. Instead, they enhance the coating of aggregates by reducing surface energy at the aggregate/binder interface and/or decreasing internal friction (16). The latter, foaming, can be accomplished by introducing atomized water directly into the hot binder or by incorporating microparticles, such as zeolites or water-attracting fillers, which harbor unleashed water during the production process of the mixture at temperatures typically exceeding 120°C (17-19).

The aim of this study is to analyze the impact of incorporating 3 multi-source, industrial waste-based, and water-releasing products in the manufacture of WMA with the goal of reducing costs and consumption of virgin materials involving its production while achieving comparable or superior mechanical properties to an HMA. The fundamental selection criteria were to find potential recycled materials that were compatible with the bitumen and could manage to provide water to achieve the indirect foaming of the binder. Additionally, another important feature was the absence of leachates that could potentially harm the overall performance of the mixture. The first considered material is a zeolite by-product that is a commonly used catalyst applied during the refining process of petroleum. The second and third analyzed waste additives are products that come from rapid pyrolysis processes of biomass or, a mixture of plastic with biomass. The three products were chemically characterized, and evaluated by analyzing their water-retaining and water-releasing capacity to meet their exigencies to be used as water-containing additives for WMA production. After the selection, the design and performance of the three modified WMA were compared with a conventional HMA, while all sharing the same aggregate skeleton as well as the same manufacture and compaction temperature.

2. MATERIALS AND METHODOLOGY

2.1. Materials

Within this research, three distinct waste-based additives from various origins underwent analysis: a zeolite by-product (referred to as Additive 1), employed in the fluid catalytic cracking (FCC) stage during the petroleum refining procedure to expedite the breakdown of dense molecules. This is facilitated by its crystal structure, which offers channels and chambers for enhancing molecular interaction (20, 21). The second additive (Referred to as Additive 2) is a derived product from rapid pyrolysis processes of charred acacia tree splinters, and the third one (Referred to as Additive 3) is a by-product from charred plastic. Since the analyzed materials are derived from waste, their granulometry is variable and cannot be controlled, nonetheless, their particles were sieved and pass through the 1 mm sieve, having a higher concentration of particles smaller than 0.063 mm, especially in that of the zeolite by-product which is the finest material. Figure 1 exhibits the physical appearance of the materials.

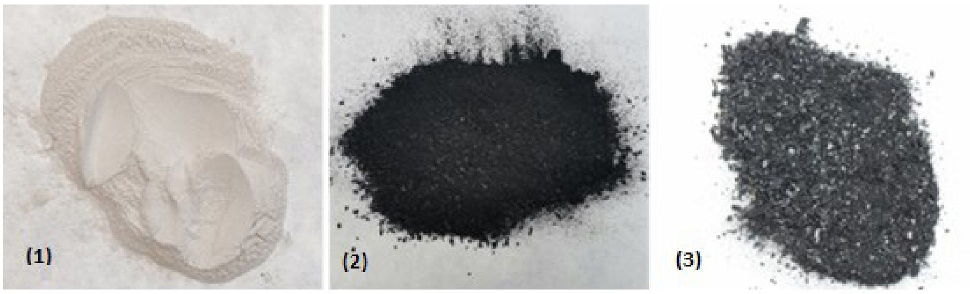

FIGURE 1. Physical appearance of additives (1: Additive 1; 2: Additive 2; 3: Additive 3).

The three additives have a maximum water absorption capacity of approximately 15% of their overall weight, although their natural humidity is highly dependent on storage conditions and duration. Additive 1 reached its maximum absorbed humidity after about 400 hours, whereas Additive 2 and Additive 3 took 50% longer to achieve similar values in a climate-controlled chamber with a humidity level of no less than 98%. For the laboratory tests on the bituminous mixtures, however, the three by-products were conditioned to achieve a uniform water content of 10%.

Regarding their chemical composition, Additive 1 is primarily composed of aluminum oxide (Al_2O_3), making up 45.522% of its composition, with traces of rare earth elements (REEs) and phosphorus pentoxide (P_2O_5), both present in quantities below 1.3%. Its most abundant metallic elements are nickel, vanadium, and magnesium. Additive 2 contains a high percentage of carbon, its most abundant element, accounting for 84.200% of its composition. This is consistent with its biomass origin. Other significant elements in this material include hydrogen (2.760%) and iron (6.870%), along with potassium (2.257%). For Additive 3, carbon is the most abundant element, comprising 37.450% of its composition, followed by smaller amounts of hydrogen (1.960%) and nitrogen (1.290%), which aligns with its polymeric origin. Among its metallic components, it has a substantial presence of calcium, silicon, aluminum, and iron (12–17%), while phosphorus, sodium, and chlorine are present in smaller quantities (1.5–3.8%). Some of the chemical components (Carbon, hydrogen, and nitrogen) of the additives were determined using the instrumental method UNE-CEN/TR 17674:2022-IN, while metallic elements were determined and quantified by following the UNE-EN 24883:1993 norm.

2.2. Methodology

To assess the viability and the effect of using the three indirect foaming additives, comparisons with a traditional AC-22 asphalt mix (EN 13108-1) (22) were conducted. The mineral aggregates consisted of limestone of three size fractions according to the asphalt mixture requirements, while the bitumen utilized was a conventional B 50/70, and the optimal asphalt dosage determined was 4.5% of the overall weight of the mixture. The main physical characteristics of the reference mix are displayed on Table 1.

Table 1. Main physical characteristics of the reference HMA.

Properties	AC22 50/70
Optimal bitumen content (% over mix weight)	4.5
Bulk density (Mg/m3), EN 12967-6	2.528
Air void content (%), EN 12967-8 4.1	4.7
Filler/bitumen ratio	1.1
Mix temperature (ºC)	160

After the reference mix was established, the three WMA mixes were modified as with a dry additive by replacing a fraction of the aggregates, 1% of the calcium carbonate filler for all the cases, with the evaluated products to aim to reduce the manufacturing temperature from 160°C to 140°C. The compaction temperature was also set to be the same as that of the mixture. Thus, in this study the 4 considered mixes are labeled as follows: HMA-R (reference), which was manufactured at the standard temperature of HMA; WMA-1 (manufactured with Additive 1, zeolite-based); WMA-2 (produced with Additive 2, charred acacia) and WMA-3 (Made with Additive 3, charred plastic). Table 2 shows the general composition of the 4 analyzed mixtures.

Table 2. Composition of analyzed mixes.

Mixture	18/25 mm limestone	18/25 mm limestone	18/25 mm limestone	Calcium carbonate Filler	Waste additive 1	Waste additive 2	Waste additive 3	Asphalt binder type	Asphalt binder content	Mixture Tª (ºC)
HWA-R	20	30	45	5	0	0	0	B50/70	4.5	160
WMA-1	20	30	45	4	1	0	0	B50/70	4.5	140
WMA-2	20	30	45	4	0	1	0	B50/70	4.5	140
WMA-3	20	30	45	4	0	0	1	B50/70	4.5	140

The mechanical performance of the three modified WMA formulations was compared with that achieved by conventional HMA to assess the real feasibility of utilizing these multi-source waste additives in the production of WMA for road construction. Traditional Marshall samples were prepared for the four analyzed mixes in accordance with the specifications for the preparation and manufacture of the test specimens outlined below. In this respect, the conducted tests were the stiffness modulus at a temperature of 20°C (EN 12697-26 annex C) (23), water sensitivity test (EN 12697-12) (24) and triaxial test (EN 12697-25 method B) (25) both at 60°C.

3. RESULTS AND DISCUSSION

Table 3 displays the physical properties of the studied mixes. Addition of additives 2 and 3 resulted on an increase of the air void content (26), compared to the reference mix, with values of 5.4 and 6.1 respectively, while the additive 1 reached a lower value of 3.9. The zeolite additives (as the one depicted as Additive 1) typically enhances the malleability of a bituminous mixture at lower temperatures and facilitates its compaction. Overall, these results are consistent with the literature, showing a lower percentage of voids compared to the reference mixture (16, 20) (which also could be helped by the finer granulometry of this material). Consequently, the Bulk density (27) of the HMA-R and the WMA-1 are very similar, while WMA-2 and WMA-3 exhibit lower values (This suggests that both Additives 2 and 3 are less effective than Additive 1 in reducing the manufacturing temperature of asphalt mixtures, especially considering that AC-22 is a dense-graded mixture and that achieving similar volumetric parameters and a mechanical behavior comparable to that of HMA is a desired outcome). The physical appearances of one of the modified mixes and the reference mix are depicted in Figure 2, illustrating that the aggregates exhibit appropriate coating, and the manufacturing process is entirely normal.

The mechanical performance of WMA, produced at 140°C with 1% of the chosen by-product additives, is compared to the outcomes of the reference HMA fabricated at 160°C. Figure 3 portrays the results of the water sensitivity test conducted on the analyzed mixtures. The figure presents the indirect tensile strength for both dry and saturated subsets of specimens, along with the ratio of restrained strength (ITSR: indirect tensile strength ratio). The results show that the WMA manufactured with Additives 2 and 3 presented considerably lower values of ITSR index (54.4% for Additive 2 and 61.2% for Additive 3) than the traditional HMA (with 89.0%), as well as a significant reduction in mixture ITS. Nevertheless, the findings suggest that WMA-1 attained tensile strength values similar to those of the HMA-R mix for both wet-conditioned and dry subsets. Furthermore, this mixture exhibited a minor reduction in moisture resistance influence, while being able to fulfill the most exigent criteria for these types of materials in Spanish specifications for the construction of roads (28). Additionally, a visual inspection of the specimens after testing indicated that, in all the analyzed mixtures, the aggregates displayed proper bitumen coating in the dry subset. However, in the saturated subset, WMA-2 and WMA-3 showed inadequate coating, signifying that these additives, two and three, negatively impact the adhesion between the bitumen and aggregates under water action.

Figure 2. (Left) Reference mix-HMA-R (Left-center) Modified mix-WMA-1(Right-center) Modified mix-WMA-2(Right) Modified mix-WMA-3.

Table 3. Physical properties of analyzed mixes.

Properties	HMA-R	WMA-1	WMA-2	WMA-3
Optimal bitumen content (% over mix weight)	4.5	4.5	4.5	4.5
Bulk density (Mg/m3), EN 12967-6	2.528	2.524	2.426	2.435
Air void content (%), EN 12967-8 4.1	4.7	3.9	5.4	6.1
Filler/bitumen ratio	1.1	1.1	1.1	1.1
Mix temperature (°C)	160	140	140	140

Figure 4 illustrates the stiffness modulus of WMA with the by-product additives alongside the reference HMA. Results show that there is a decrease in the stiffness modulus for all three modified mixes (This outcome aligns with expectations from the literature for this type of WMA technology, particularly in mixtures produced with a temperature reduction of 20°C, as is the case in this study) (29). The results present that both WMA modified with Additive 1 and Additive 2 show comparable values with a decrease of about 20% to those of HMA-R stablished at 5586 MPa, with 4531MPa and 4427MPa correspondingly. In contrast, WMA-3 displayed significantly lower stiffness, at 3374 MPa, representing a 40% reduction, double the decrease detected with Additives 1 and 2.

While analyzing the triaxial test results, the WMA modified with Additive 1 presents virtually the same result, 1.42%, of final deformation as the reference HMA. However, mixes modified with additives 2 and 3 show a higher final deformation with values of 1.65% and 1.79% each, which is in consonance with some authors (30) who state that WMA with by-products additives could cause a higher susceptibility to deformation. On Figure 5 the results of final deformation of the 4 studied mixtures after triaxial test are displayed.

These results, showcasing the behavior of the analyzed mixes, provide practical guidance for the asphalt industry by emphasizing the importance of additive selection in WMA production. By meeting strict specifications—such as those outlined in Spanish road construction standards—and delivering environmental benefits, Additive 1 supports the adoption of sustainable practices without sacrificing pavement quality. These findings bridge a critical knowledge gap, offering data and analysis that support the adoption of WMA in regions where it is not yet widely practiced. Demonstrating that WMA can meet or exceed the performance standards of HMA further strengthens the case for its global implementation.

The results also contribute to the existing body of literature by providing empirical data on the performance of WMA with specific by-product additives at reduced temperatures. This enhances the understanding of the influence of different additives on the mechanical properties and durability of asphalt mixtures under laboratory conditions.

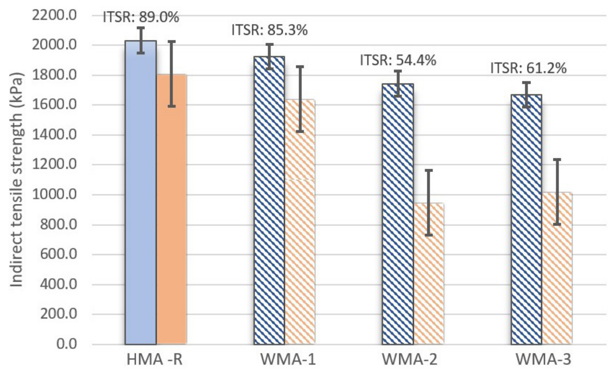

FIGURE 3. Results of the water sensitivity test for HMA and WMA with by-product additives.

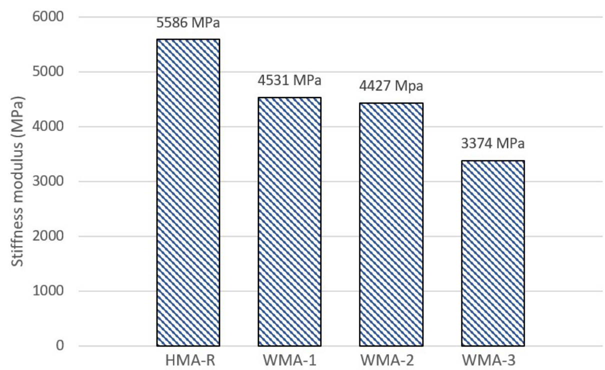

FIGURE 4. Stiffness modulus for both HMA and WMA with Additives.

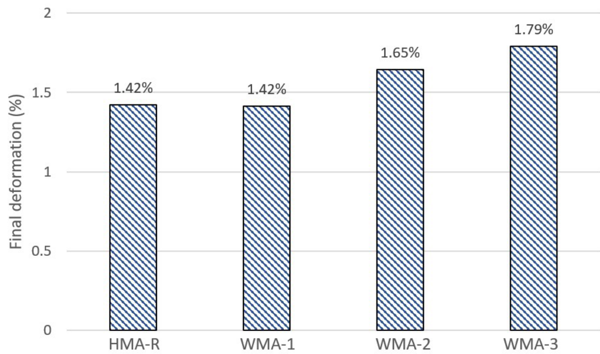

FIGURE 5. Final deformation values of WMA modified with By-products additives.

4. CONCLUSIONS

This study assessed the feasibility of incorporating industrial waste by-products, sourced from recycled materials, to foster a circular economy and/or industrial symbiosis in local areas through the production of Warm Mix Asphalt (WMA) via the indirect bitumen foaming process. Three different by-products/wastes were analyzed: a zeolite by-product (Additive 1), a derived product from the rapid pyrolysis of charred acacia tree splinters (Additive 2), and a by-product from charred plastic (Additive 3). Based on the evaluation of mechanical performance in WMAs incorporating these by-products, as well as in comparison to a conventional Hot Mix Asphalt (HMA) sharing the same mineral skeleton and binder content, the following conclusions were drawn:

- Additive 1 (zeolite-based by-product) effectively reduced the manufacturing temperature of asphalt mixtures from 160°C to 140°C without compromising mechanical or volumetric properties. This represents a significant advancement, confirming that alternative additives can maintain or enhance asphalt performance at lower production temperatures, leading to energy savings and reduced greenhouse gas emissions. Additive 1 demonstrated superior compaction characteristics, as evidenced by its lower air void content (3.9%) and bulk density comparable to the reference HMA. Mechanical tests revealed indirect tensile strength (ITS) values similar to HMA-R for both dry and saturated specimens, while the indirect tensile strength ratio (ITSR) met the stringent criteria specified in Spanish road construction standards. The stiffness modulus and triaxial test results closely aligned with those of HMA-R, confirming that Additive 1 preserved the mechanical integrity of the mixture.
- Additives 2 and 3, by contrast, were less effective. Mixtures incorporating these additives (WMA-2 and WMA-3) showed higher air void contents (5.4% and 6.1%, respectively) and reduced bulk densities, indicating less efficient compaction. Mechanical performance was compromised, with significantly lower ITS and ITSR values—54.4% for Additive 2 and 61.2% for Additive 3—compared to 89.0% for HMA-R, which is consistent with the results reported in the literature (31-33). The stiffness modulus decreased by approximately 20% for WMA-2 and 40% for WMA-3, while triaxial tests revealed increased susceptibility to deformation. These

findings, supported by visual inspections post-moisture conditioning, suggested that Additives 2 and 3 adversely affect bitumen-aggregate adhesion and overall performance under water exposure.
- The successful application of Additive 1 aligns with global efforts to reduce the carbon footprint of construction activities. By lowering production temperatures without compromising asphalt mixture quality, Additive 1 promotes more sustainable infrastructure development while contributing to significant energy savings and reductions in CO_2 emissions.

By replacing virgin materials with waste-derived additives, reducing energy consumption, and lowering greenhouse gas emissions, Additive 1 represents a promising step towards industrial symbiosis and the circular economy in road construction. Its adoption could yield substantial financial and environmental benefits for the asphalt industry, encouraging the use of innovative, eco-friendly materials in future road-infrastructure projects.

Authorship contribution statement

Walter Mera: Data curation; Formal analysis; Validation; Visualization; Writing – original draft.
María Carmen Rubio Gámez: Conceptualization; Funding acquisition; Investigation; Methodology; Project administration; Resources; Supervision; Validation.
Fernando Moreno Navarro: Conceptualization; Data curation; Formal analysis; Funding acquisition; Investigation; Methodology; Project administration; Resources; Supervision; Validation.
Gema García Travé: Data curation; Formal analysis; Investigation; Validation.
María del Mar Colas: Conceptualization; Data curation; Formal analysis; Funding acquisition; Investigation; Methodology; Project administration; Supervision; Validation.
María González González: Conceptualization; Formal analysis; Funding acquisition; Investigation; Methodology; Supervision; Validation.

Declaration of competing interest

The authors of this article declare that they have no financial, professional or personal conflicts of in-terest that could have inappropriately influenced this work.

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