



Influence of chemical additives and wax modifiers on odor emissions of road asphalt

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HIGHLIGHTS

- Use of artificial olfactory system approach to measure asphalt emission odor.
- Assessment of a specific odor fingerprint of asphalt and asphalt additives.
- Temperature as a key parameter in the generation of specific odors in HMA e WMA production.
- Detection of no changes in asphalt odor emissions after introduction of different additives.

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ABSTRACT

The use of a broad range of asphalt additives is a well-established practice in road pavement engineering for the production of high-performance hot-mix and warm-mix asphalt mixtures. The study aimed to verify and to assess, through an analytical-sensory approach based on artificial olfactory system (AOS), the effects of five different asphalt additives (chemical additives, odor suppressant agent and wax modifiers) on the odorous patterns of asphalt emissions at typical mixing and laying temperatures. The AOS has made possible to identify a specific odor fingerprint of each additive. However, once added to asphalt, these agents did not establish with binder effects of synergy, additivity or antagonism, but appear to be as neutral elements by an odorous point of view. The odorous patterns of emissions generated by heating neat asphalt at various temperatures in laboratory scale tend to coincide with those of asphalt/additive mixtures, underlining how the bituminous binder odor resulted to be hiding or masking compared to that of only-additives.

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1. Introduction

The primary requirement of road operators to contain pavement construction and maintenance costs, while extending at the same time the pavement service life, continues to stimulate the asphalt industry to research and to develop increasingly efficient technologies and solutions. One of the most popular and cost-effective measure to potentially overcome these challenges is represented by the use of a broad range of asphalt additives, specifically tailored for various applications in different climates and kinds of asphalt base material [1,2].

Several chemical modifiers or bio-based oils, such as extenders, antioxidants, hydrocarbons, anti-stripping agents and adhesion

promoters, have been formulated to boost and optimize specific binder or bituminous mixture characteristics [3–5]. An increasing international awareness on environmental sustainability, in terms of both limitation of non-renewable natural resources use and substantial reduction in energy consumption and in emissions released into the atmosphere, has led to the development of two different families of products. On the one hand, with the growing interest in increasing the content of recycled aggregates, recycled asphalt pavement (RAP) and recycled asphalt shingle (RAS) or industrial by-products (tires, steel slags, glass), in bituminous mixtures several recycling agents (RAs) have been formulated. RAs are classified as two types: rejuvenating agents and softening agents [6,7]. On the other hand, specific additives or modifiers have been studied to lower temperatures at which the bituminous mixtures are mixed and placed on the road pavement, while maintaining the demanding road specifications. This technology, known as

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warm mix asphalt (WMA), uses production and compaction temperatures between 90 and 140 °C with a decrease up to 30 °C compared to traditional hot-mix asphalt (HMA), reducing asphalt fumes, plant airborne emissions and burner fuel demands [8]. The WMA technology can be basically grouped in three main classes: organic waxes (workability promoters), chemical additives and foaming technology [9,10].

Several literature experiences have deeply examined the effectiveness and the efficacy of these additives, but no study has analyzed the impact that the addition of these agents causes on the odorous emissions generated during storage and handling of additive asphalt at high temperatures. Nowadays asphalt and asphalt operations face increasing public concern about odor emissions, so much that EAPA (European Asphalt Pavement Association) in Europe and NAPA (National Asphalt Pavement Association) in the USA have set up in recent years technical discussions to address this strategic issue [11]. Although the HMA plants have implemented several measures in accordance to the best available techniques (BAT) for reducing emissions, such as placement of higher stacks, implementation of full-scale fabric filter or bio-filter, wet and chemical scrubbers, activated carbon absorbers and covering the storage areas [12], asphalt odor emissions remain a cause of annoyance and nuisance to workers and neighborhoods. The airborne asphalt emissions are a complex mixtures hydrocarbon aerosols, vapors and gases, generally composed by product of combustion, volatile organic compounds (VOCs) and polycyclic aromatic hydrocarbons (PHAs) [13,14]. The odor of these gaseous complex mixtures varies with a number of factor, including source characteristics, atmospheric dispersion and above all receptor sensitivity. The highly subjective nature of odor is also testified by its scientific definition: organoleptic attribute perceptible by the olfactory organ on sniffing certain volatile substances (ISO 5492), in other words the sensory response to the inhalation of air containing chemicals [15]. Thus, an objective measurement of odor in the conventional sense can be very difficult. There are two traditional approaches for measuring odors, i.e. instrumental evaluation (chemical quantification) and sensory evaluation (olfactometry), which imply different types of drawbacks [16]. A new frontier in the odor objectification is represented by an instrument which is designed to mimic the human sense: the artificial olfactory system (AOS) or electronic nose [17,18]. It is an electronic device that uses a sensor array, whose resulting electronic response is processed by a suitable pattern-recognition (PARC) system capable of producing a two-dimensional spectral pattern (fingerprint or smellprint) that is specific for a particular mix of chemical compounds [19,20]. Although the AOS approach is a common practice in various industrial sectors, their potentials in asphalt industry have only recently been investigated with positive results [21,22].

On the basis of these considerations, this study aimed to verify and to assess, though an analytical-sensory approach based on AOS technology, the effects that some asphalt additives cause to the odorous patterns of asphalt emissions at typical HMA and WMA process temperatures. At the same time, the potential of an odor suppressant agent, which is specifically formulated for asphalt with the ambition to act as odor neutralizer, was investigated.

2. Materials characterization

Two unmodified 70/100 penetration grade asphalts produced in two different plants refining Arabian crude oils, hereinafter referred to as B1 (Pen_{25 °C} = 84 0.1 mm; R&B = 46.4 °C) and B2 (Pen_{25 °C} = 86 0.1 mm; R&B = 46.2 °C), were used in the experimental program. From the two virgin asphalts, five binary asphalt/oil or asphalt/wax blends were prepared adding a predetermined concentration of each additive to asphalt. Specifically, three liquid oil products were used, i.e. two chemical additives, commercially

considered as adhesion enhancers (CA1 and CA2) and an odor suppressant additive (OSA). Two different types of wax, which are workability promoters, were also tested: Asphaltan B[®] – Montan wax with Clariant fatty acid starch (MW) and Sasobit[®] Sasol wax (SW). The chemical additive CA1 is a compound consisting of amine derivatives, CA2 is a complex mixture of vegetable origin added with enhancer substances and OSA consists of a complex mixture of hydrocarbons, alcohols and essential oils. MW is a semi-synthetic refined Montan-type wax, which is derived by solvent extraction of fossil deposits of lignite-rich or brown coal vegetable substances, blended with fatty acid amide. SW is a synthetic hydrocarbon hard wax obtained as by-product of the Fischer-Tropsch (FT) process, where hydrocarbons are synthesized from hydrogen and carbon monoxide. Both pure waxes show a melting point, also defined congealing point, approximately equal to 100 °C and solidify in asphalt between 70 and 115 °C [23,24]. Asphalt/wax mixtures were obtained by adding 3% by weight of wax to the asphalt, whereas liquid additive amounts (by weight of asphalt) were 0.50% for the additive CA1, 0.75% for CA2 and 0.017% for OSA. The dosage of the selected additive was chosen based upon manufacturer's recommendation and the literature suggestions. Each additive or modifier was incorporated into preheated asphalt at 160 ± 5 °C, homogenizing the mixture for 15 min at adequate stirring.

In order to preliminary assess the effects of additives on physico-chemical properties of asphalt, separation and detection of SARA components (acronym for saturates, aromatics, resins and asphaltenes) and rheological testing were carried out both on asphalts and on asphalt/additive mixtures.

The SARA fractions were identified using the layer chromatography-flame ionization detection (TLC-FID) technique with the Iatroscan MK-5: TLC-FID analyzer (Iatron Laboratories Inc.), according to IP 469/01:2006 standard. The separation was achieved by using a three-stage solvent development sequence: saturates were eluted with heptane, aromatics with a solution of toluene and heptane (80:20 by volume) and resins with a solution of dichloromethane and methanol (95:5 by volume).

The SARA numerical values (Table 1) describe how addition of oils and waxes caused a variation in the asphaltene fraction, with a consequent re-adjusting of the others. The trend resulted to be more marked for the wax modified asphalts. This behavior may be attributable, in addition to a possible short-term asphalt binder aging which could have occurred during the additives blending at 160 °C, to the insolubility of waxes and the incomplete miscibility of oils in the solvents used for the fractions separation procedure.

Anton Paar Physica MCR 101 dynamic shear rheometer (DSR) was used, in accordance to EN 14770:2012, to measure the rheological properties of asphalt/additive blends in terms of complex shear modulus ($G^* = \tau_{\max}/\gamma_{\max}$), phase angle (δ) and shear viscosity ($\eta = \tau/\dot{\gamma}$). G^* can be considered the sample's total resistance to deformation when repeatedly sheared, whereas δ is defined as

Table 1
Asphalt and asphalt/additive SARA fractions: mean and standard deviation (n = 20).

Blend	Saturates [%]	Aromatics [%]	Resins [%]	Asphaltenes [%]
B1	5.2 (0.4)	52.6 (2.7)	26.6 (1.7)	15.6 (1.2)
B1-CA1	5.4 (0.6)	46.1 (2.2)	29.9 (1.4)	18.6 (0.8)
B1-CA2	5.4 (0.6)	50.5 (3.2)	27.2 (2.2)	17.0 (0.9)
B1-OSA	5.2 (0.6)	47.7 (2.9)	27.3 (1.9)	19.7 (1.7)
B1-MW	5.9 (0.2)	45.1 (2.7)	27.7 (1.3)	21.3 (1.7)
B1-SW	5.1 (0.4)	48.3 (2.1)	26.4 (1.5)	20.2 (1.1)
B2	2.3 (0.2)	55.3 (2.0)	25.5 (1.8)	16.9 (0.7)
B2-CA1	2.5 (0.3)	48.9 (2.5)	27.6 (1.4)	20.9 (0.8)
B2-CA2	2.8 (0.4)	47.0 (2.2)	28.6 (1.8)	21.7 (1.3)
B2-OSA	3.0 (0.4)	47.8 (2.2)	30.5 (1.2)	18.7 (1.3)
B2-MW	2.4 (0.3)	47.3 (2.2)	26.6 (1.3)	23.9 (1.5)
B2-SW	2.8 (0.4)	47.1 (2.3)	27.2 (1.4)	22.9 (1.2)

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