

The Use of Zinc Oxide in Asphalts: Review

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Abstract: Zinc oxide (ZnO) is a wide-gap semiconducting material which is chemically stable at high temperatures and has been shown to be compatible with asphalt binders. Additionally, semiconducting nanoparticles such as ZnO could help to improve urban air quality. This has encouraged the use of this material as a binder and asphalt mix modifier. A review on the use of ZnO as an asphalt binder modifier was conducted in this study. Based on the review which we carried out, the following were mainly synthesized and described: the content of ZnO as a modifier, the mixing processes of the binder and the ZnO, the manufacturing process of the modified mix, the type of asphalt binder and/or modified mix, the tests carried out, general conclusions, and environmental effects. ZnO micro-particles increase the aging resistance of the asphalt binder (mainly to ultraviolet radiation (UV)). ZnO tends to increase rutting resistance and adhesion with aggregates, improving resistance to moisture damage. It also tends to improve binder fatigue resistance. At low service temperatures, the performance of ZnO as a modifier is unclear, and further studies should be performed. Few studies have evaluated the effect of ZnO on the environment when it is used as an asphalt binder modifier. Future studies should evaluate the effect of this technology on the environment, perform economic analyses, evaluate the physical–chemical interaction of the modified binder with the aggregate, and measure the long-term properties of asphalt mixtures. ZnO is viewed as an environmentally friendly material and as a promising modifier of asphalt binders for pavement construction.

Keywords: asphalt; binder; modifier; zinc oxide (ZnO); nano-ZnO



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1. Introduction

Zinc is the fourth-most widely used metal in the world after iron, aluminum, and copper [1]. Zinc oxide (ZnO) is the main chemical product produced from metallic zinc. Nano-zinc oxide (nano-ZnO) is produced from the synthesis process of an aqueous solution of zinc nitrate (Zn(NO₃)) and hexamine (C₆H₁₂N₄) with some reagents such as polyethylene glycol at a temperature above 90 °C [2]. ZnO powder has traditionally been used as a white pigment and as an additive for rubber. Its use is estimated to be between 50% and 60% in the rubber industry (as a vulcanization activator and improving thermal conductivity). The second-largest application is in ceramics [3]. This is mainly because ZnO is a material with high heat capacity, high conductivity, and thermal and chemical stability [4]. It is also a material with a low coefficient of expansion and optical ultraviolet (UV) absorption [5]. In the concrete industry, its addition in small quantities to Portland cement can delay setting and hardening [6]. It can also improve the water resistance of concrete [7]. ZnO is also used in plastics, pigments, and coatings; in cosmetic, medical, and dental applications; in fertilizers, animal feed, and dietary supplements; and in electronics, optoelectronics, glass, and lubricants, etc. [5,8–10].

Zinc oxide (ZnO) is a wide-band gap semiconductor material with piezoelectric properties [11,12]. These properties, in conjunction with its nanometer size, have attracted the interest of numerous researchers in the field of modified asphalt to improve UV aging

resistance [2,13–17]. It is a high-refractive-index material, as it can be an electrical conductor when properly doped, and is thermally stable at extremely high temperatures (at least ~ 1800 °C) [5]. Piezoelectric crystals of ZnO can transform mechanical energy into an electrical signal, or vice versa [18].

ZnO is generally classified as a colorless and low-toxicity material that does not cause skin or eye irritation; additionally, there is no evidence of carcinogenicity, genotoxicity, or toxicity in humans [19,20]. However, the dust can be hazardous by inhalation or ingestion [21], as it causes a condition known as zinc fever [5]. The current evidence shows that ZnO particles and nanoparticles do not penetrate skin cells and remain in the outer layer of undamaged skin (the stratum corneum) with low systemic toxicity [22]. ZnO is a material that can act as a catalyst in redox reactions that promote the photodegradation of pollutants [23].

Some aspects that motivate its use in binders and asphalt mixtures are as follows: (i) it is a material with high compatibility with asphalt and is stable at high temperatures [24–26]; (ii) it improves the UV aging resistance of asphalt [14]; (iii) adding nano-ZnO to asphalt mixtures reduces the acid component and increases the basic component of the binder, improving its adhesion with the aggregate [27]; (iv) photocatalytic infrastructures based on semiconducting nanoparticles such as ZnO can be promising solutions with which to improve urban air quality due to their ability to mineralize toxic organic compounds; (v) car exhaust components such as ozone (O_3), cyclooxygenase (COX), volatile organic compounds (VOCs), nitrogen oxides (NOX), carbon dioxide (CO_2), and nitric oxide (NO) emissions tend to decrease in presence of ZnO surfaces [28,29]; (vi) ZnO could remove acidic contaminants such as nitrate (NO_3) and sulfate (SO_4) from the water rainfall on the pavement surface [30].

The main objective of this research was to carry out a review regarding the use of ZnO as a binder and asphalt mix modifier. This review may be used by pavement researchers around the world as a starting point for future studies on the subject. Additionally, it will be a source of reference for researchers, academics, and students in the areas of civil engineering, pavements, roads, geotechnical engineering, materials, and related fields. As important information, it should be noted that only one study has reported a review on this subject, which can be consulted in Debbarma et al. [2]. However, in that study, the central topic was the use of different nano-materials in asphalt mixes, and the information reported on ZnO was scarce. In contrast to the study referenced in [2], this article describes more research and updates the information.

2. Methods

The consulted articles were downloaded from the following databases: ScienceDirect, Scopus, Web of Science, American Society of Civil Engineering (ASCE), Taylor and Francys, Springer, Google Scholar, and MDPI. The keywords used to download the manuscripts of interest were: “Zinc oxide + asphalt”, “ZnO + asphalt”, and “nano-ZnO + asphalt”. For the review, a specific period of time was not considered. All studies on the topic at any date of publication were reviewed.

In the present review, studies that used ZnO as a modifier of asphalt binders were considered (Figure 1). Studies were also considered in which the ZnO was previously modified to (i) improve compatibility with the polymer matrix (most polymers are hydrophobic and ZnO is hydrophilic [31]); (ii) not to lose the nanometric effect of ZnO and not to hinder its dispersion within the binder (inorganic nanoparticles have a huge surface area, a high surface energy, and agglomerate very easily [15]). Excluded from the present review were studies employing zinc, materials with high ZnO contents or zinc tailing waste as filler, ZnO as a modifier of hydraulic cements, soil stabilizers, etc. (e.g., [1,21,32–37]). Also excluded were articles in which asphalt binders were modified with ZnO-like materials, such as Zinc dialkyl dithiophosphate or zinc diethyldithiocarbamate (e.g., [38–40]).

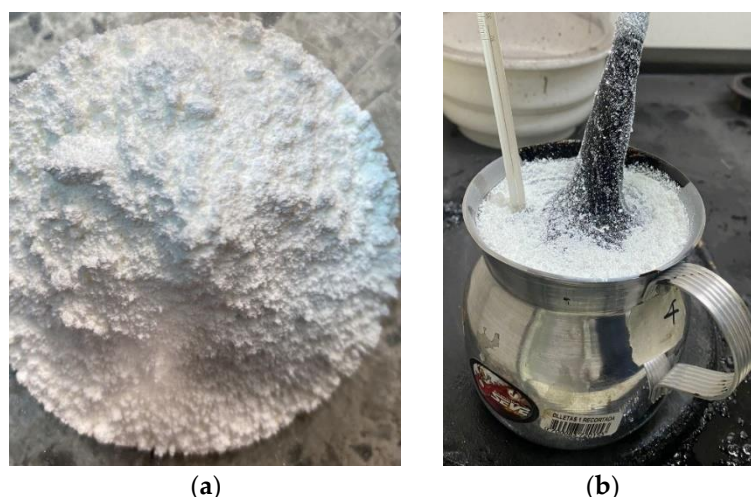


Figure 1. (a) ZnO particles; (b) ZnO as asphalt binder modifier.

The main findings of the researches are described in chronological order. The description of the studies is ordered as follows: type of asphalt binder and/or modified mix; type and content of ZnO as modifier; physical properties and tests carried out; the manufacturing process of the asphalt and/or modified mix; the main conclusions; and the environmental aspects.

Hereafter, asphalt cement (AC) with XX dmm denotes penetration (obtained at 25 °C, 100 g, 5 s; according to American Society for Testing and Materials-ASTM D5 standard [41]) of XX mm/10, and wt% refers to the percentage by weight of asphalt binder. Some acronyms that will be used later are denoted below: revolutions per minute (rpm), thin film oven test (TFOT), natural exposure aging (NEA), short-term oven aging (STOA), pressure aging vessel (PAV), rotational viscosity (RV), performance grade (PG), dynamic shear rheometer (DSR), bending beam rheometer (BBR), multiple stress creep recovery (MSCR), linear amplitude sweep (LAS), zero shear viscosity (ZSV), atomic force microscopy (AFM), Fourier transform infrared spectroscopy (FTIR), X-ray diffraction (XRD), nuclear magnetic resonance (NMR), water contact angle (WCA); Styrene–Butadiene–Styrene (SBS), fluorescence microscopy image (FMI), scanning electron microscopy (SEM), high-temperature storage stability (HTS), organic expanded vermiculite (OEVMT), tensile strength ratio (TSR), surface free energy (SFE), dynamic stability (DS), indirect tensile strength (ITS), semi-circular bend (SCB), hot-mix asphalt (HMA), stone mastic asphalt (SMA), and superpave gyratory compactor (SGC).

3. ZnO as a Modifier of Binders and Asphalt Mixtures—Review

A summary of the review which was performed herein is shown in Tables 1 and 2. Both Tables show that most of these studies evaluated the properties of binders modified with ZnO (few studies evaluated the properties on asphalt mixes; 33.3% approximately). To modify the asphalt binders, between 1 and 5 wt% ZnO was generally used. Regarding the modified binders, the most evaluated properties were conventional (penetration, softening point, etc.) and rheological. Few studies measured chemical properties. Regarding the modified asphalt mixtures, the resistance to aging and the properties at high service temperatures were mainly evaluated (few studies evaluated properties at low temperatures). ZnO tended to increase the resistance to aging, moisture damage, and rutting of binders and asphalt mixes.

Table 1. ZnO content (wt%) and tests performed.

Ref.	Asphalt Binder	ZnO Dosage (wt%)	Tests Carried out on Modified:		Tests Carried out
			Asphalt Binder	Asphalt Mix	
[16]	AC 73 dmm and AC 92 dmm	2	X		TFOT + UV aging. Penetration (25 °C), softening point, ductility (15 °C), RV (60 °C). Rheological properties (DSR). AFM and FTIR.
[17]	AC 73 dmm	3	X		TFOT + UV aging. Penetration (25 °C), softening point, ductility (15 °C), RV (60, 135 °C). HTS Test. FTIR.
[14]	AC 73 dmm	2	X		TFOT + UV aging. Penetration (25 °C), softening point, ductility (5 °C), RV (60, 135 °C). HTS test. AFM and FTIR.
[42]	AC 75 dmm	3	X		PAV aging. Softening point, penetration, ductility (15 °C), RV (60, 135 °C), XRD, FTIR, and Ultraviolet visible (UV-Vis) diffuse reflectance spectra.
[43]	AC 63 dmm	1, 2, 3, 4 and 1% OEVT	X		TFOT, PAV and UV aging. Penetration, softening point, RV (135 °C). Rheological properties (DSR).
[44]	AC 69 dmm	2, 4	X	X	Marshall, TSR, and SFE tests.
[45]	AC 69 dmm	2, 4	X	X	Marshall, TSR, SFE, and dynamic modulus tests.
[46]	AC 71, 86 and 105 dmm	1 and 3% OEVT	X		TFOT, PAV, UV, and NEA aging. Rheological properties (DSR, BBR).
[47]	AC 71, 86 and 105 dmm	1 and 3% OEVT	X		TFOT, PAV, UV, and NEA aging. Rheological properties (DSR, BBR). FTIR.
[12]	AC 64.3 dmm	2, 3, 4 and 3, 4% SBS	X	X	TFOT, PAV, UV, and NEA aging. Penetration, softening point, ductility (10 °C), and viscosity. Rheological properties (DSR, BBR). Marshall, freeze-thaw splitting, rutting, and three-point bending beam.
[28]	Does not apply. Authors used three aqueous solutions.	Does not apply. Authors used three aqueous solutions.	X	X	AFM, FTIR, and WCA. ITS.
[23]	AC 38 dmm	0.3, 0.8, 1.0% TiO ₂ + 0.2% ZnO	X		Penetration, softening point, dynamic viscosity (150 °C). Rheological properties (DSR). FTIR.
[48]	AC 62.2 dmm (PG 64-22)	1, 3, 5	X	X	RTFOT + PAV aging. Fatigue and rutting performance (DSR and BBR). RV (135, 165, 185 °C). Moisture susceptibility ITS (Lottman modified).
[49]	AC 69 dmm	1, 2, 3, 4, 5	X		TFOT + UV aging. Penetration, softening point, ductility (5 °C), storage stability. Rheological properties (DSR) and LAS tests.
[27]	AC 68 dmm	1, 2, 3	X		Penetration, softening point, flash and fire points, RV (135 °C). Rheological properties (DSR, BBR).
[50]	AC 64 dmm	0.2, 0.4, 0.6	X	X	Ductility (15 °C), Saybolt viscosity (135 °C), softening point, penetration, and specific gravity. Marshall, Immersion Marshall, ITS, boiling water, static creep (25 °C), pull-off adhesion, SCB fracture (25 °C).
[51]	AC 65 dmm	0.5, 1, 2, 3, 4	X		TFOT + UV aging. Penetration, softening point, ductility (5 °C). Rheological properties (DSR). FTIR, UV-visible-infrared-absorption and SEM observations.
[52]	AC 71 pen	3, 5, 7	X		Rheological properties (DSR)—PG, MSCR, LAS, master curve.
[53]	AC 66.5 dmm	2, 3, 4, 5, 6	X	X	RTFOT, PAV, UV aging. STOA aging. Penetration, softening point, ductility (15 °C), RV (135 °C). Rheological properties (DSR and BBR). Marshall, low-temperature indirect tensile (−6, −12, −18 °C), freeze-thaw splitting and indirect tensile fatigue (25 °C).
[54]	AC 85 dmm	nano-TiO ₂ /ZnO = 2, 4, 6 and 8wt%	X		TFOT, PAV, UV aging. SEM observations. Rheological properties (DSR, BBR)—ZSV, MSCR, LAS (25 °C) tests. FTIR.
[55]	AC 68 dmm	2, 4, 6, 8	X	X	Penetration, softening point, RV (120, 135, 150 °C), ductility. Cantabro, Marshall, ITS, Wheel Track Test (45 °C), SCB (−15, 0, 15 °C).
[56]	AC 64.3 dmm (PG 64-16)	2, 3, 4	X	X	Penetration, softening point, ductility (10 °C), RV (120 °C), DSR, BBR. Marshall, TSR, dynamic stability
[57]	It's not clear. Two AC with different wax contents	4	X		Rheological properties (DSR)—LAS, MSCR. NMR Spectroscopy.
[58]	AC 70 dmm (PG64-16) with high content of SBS 791-H (6.5wt%)	2	X		Rheological properties (DSR). FMI, ITS.
[59]	AC 60 dmm (PG 58)	3, 6, 9, 12, 15	X		Aging Index (AI). Rheological properties (DSR)—non-recoverable creep compliance, LAS (20 °C).

Table 2. ZnO influence as a modifier and optimum content (wt%) recommended.

Ref.	Resistance to:						Adherence	Optimum Content of ZnO (wt%)
	Rutting	Fatigue	Temperature Cracking		Moisture	Aging		
			Intermediate	Low				
[16]	-	-	-	-	-	I	-	-
[17]	-	-	-	-	-	I	-	-
[14]	-	-	-	-	-	I	-	-
[42]	-	-	-	-	-	I	-	-
[43]	-	-	-	-	-	I	-	1% OEVMT + 3% ZnO
[44]	-	-	-	-	I	-	I	-
[45]	-	-	-	-	I	-	I	-
[46]	I	-	-	I	-	I	-	-
[47]	I	-	-	I	-	I	-	-
[12]	I	I	-	I	I	I	-	-
[28]	-	-	-	-	S	-	S	-
[23]	-	-	-	-	-	I	-	-
[48]	I	D	-	-	I	I	-	5
[49]	I	I	-	-	-	I	-	3
[27]	I	I	-	I	-	-	-	3
[50]	I	-	-	-	I	-	I	0.6
[51]	I	-	-	-	-	I	-	3
[52]	I	I	-	-	-	I	-	7
[53]	I	I	-	I	I	-	-	5
[54]	I	I	I	-	-	I	-	nano-TiO ₂ /ZnO = 4
[55]	I	D	-	-	-	-	I	6
[56]	I	S	-	S	I	I	-	-
[57]	I	I	-	-	-	I	-	-
[58]	I	I	-	-	-	I	-	-
[59]	I	-	-	-	-	I	-	-

I: Increase; D: decrease; S: similar.

The following is a summary of the information consulted chronologically.

Du et al. [16] evaluated the effect of nano-ZnO (15–25 nm) on the morphology and UV aging properties of two asphalt binders (AC 73 dmm and AC 92 dmm). Both AC were mixed with nano-ZnO (2 wt%) at 150 ± 5 °C at 5000 rpm for 60 min using a high shear mixer. Then, mixing was continued using a paddle agitator at 2000 rpm for 90 min. This same process was copied for the ACs without the nano-ZnO for comparison. The asphalt binders were aged in TFOT + UV. The TFOT residue was aged with UV for a period of 0 to 24 days in an oven with a 500 W UV lamp. The UV radiation intensity was 8 W/m^2 . The penetration (25 °C), softening point, ductility (15 °C), RV (60 °C), and rheological properties were measured on the binders before and after aging using a DSR. Tests were also carried out using AFM and FTIR. The aging resistance of both binders improved, but the effect of nano-ZnO depended on the type of base binder.

Liu et al. [17] modified an AC 73 dmm with 3 wt% nano-ZnO (15–25 nm). The surface of the nanoparticles was previously modified with γ -methacryloyloxypropyltrimethoxysilane (MTS), 3-aminopropyltriethoxysilane (APTS), and γ -(2,3-epoxypropoxy) propyltrimethoxysilane (EPTMS). Mixing was performed at 150 ± 5 °C, 5000 rpm for 60 min, using a high shear mixer. Then, mixing was continued with a paddle stirrer at a speed of 2000 rpm for approximately 90 min. Researchers performed the HTS Test; aged the binders in TFOT + UV; and performed softening point, penetration, ductility (15 °C), RV (60, 135 °C), and FTIR tests. The resistance to UV aging improved with nano-ZnO, and the resistance changed depending on the type of modifier used on its surface. According to the researchers, surface modification of nano-ZnO was an effective method with which to improve the binder's resistance to UV aging.

Zhang et al. [14] modified an AC 73 dmm with nano-ZnO (15–25 nm) and nano-ZnO modified on the surface with c-(2,3-epoxypropoxy) propyltrimethoxysilane. Mixing of AC with 2 wt% nano-ZnO (without and with modification) was performed at 150 ± 5 °C at 5000 rpm for 60 min using a high shear mixer. Then, mixing was continued using a paddle agitator at 2000 rpm for 90 min. The same process was performed on unmodified AC. The

AC (unmodified and modified) was aged at TFOT + UV. The researchers performed softening point, penetration, ductility (5 °C), RV (60 and 135 °C), FTIR, AFM, and HTS tests. The compatibility between AC and nano-ZnO improved along with nano-ZnO surface modification. The UV aging resistance improved when AC was modified with nano-ZnO, and was higher when the latter was modified with c-(2,3-epoxypropoxy) propyltrimethoxysilane. On the other hand, in the same year, Zhang et al. [15] published a similar study using two other nanoparticles (nano-SiO₂ and nano-TiO₂). The conclusions in the latter study were the same as those in the former. However, Zhang et al. [15] concluded that, compared to the other two nanoparticles, the nano-ZnO-modified binder showed the best resistance to UV aging after surface modification.

Zhang et al. [42] modified an AC 75 dmm with nano-ZnO (15–25 nm) and OEVMt (sieve 300). The nano-ZnO surface had been previously modified with 3-aminopropyltriethoxysilane. The content of both nanomaterials was 3wt%; however, the individual content of nano-ZnO and EVMt was unclear. Mixing was performed at 150 ± 5 °C and 5000 rpm for 60 min using a high shear mixer. It was then mixed using a paddle stirrer at a speed of 2000 rpm for 90 min. The same process was also performed for the base binder. The modified binders were aged in PAV, and softening point, penetration, ductility (15 °C), RV (60, 135 °C), XRD, FTIR, and Ultraviolet visible (UV-Vis) diffuse reflectance spectra tests were performed. The nano-ZnO improved the resistance to UV aging.

Zhang et al. [43] modified an AC 63 dmm with 1 wt% OEVMt and 1, 2, 3, and 4 wt% nano-ZnO (average particle size 20 nm, which was previously surface-modified with c-(2,3-epoxypropoxy) propyltrimethoxysilane). Mixing was carried out at 150 °C at 4000 rpm for 60 min using a high shear mixer. Then, mixing was continued at a rotational speed of 2000 rpm at 150 °C for approximately 90 min. The same preparation process was also performed on the base binder. The binders were aged in TFOT, PAV, and UV. Penetration, softening point, RV (135 °C), and DSR tests were performed on the samples. OEVMt and nano-ZnO improved the resistance to thermal oxidation and aging. The recommended content of these materials was 1% OEVMt + 3% nano-ZnO. Nano-ZnO has a negative influence on the physical properties of the binder when its content exceeds 4 wt%.

Hamidi et al. [45] modified an AC 69 dmm with nano ZnO (2 and 4 wt%). The AC was mixed with nano ZnO for 4–5 min at 130–140 °C and 14,000 rpm. The researchers manufactured asphalt mixes designed by the Marshall procedure and measured the TSR and SFE on the samples. Nano ZnO decreased the acidity of the asphalt binder, improving the adhesion with an acid aggregate such as granite. It increased the SFE of adhesion between the aggregate and asphalt. This increased the moisture damage resistance of the asphalt mix. A similar study with the same conclusions can be found in Hamedi and Nejad [44]. However, in the latter study, the researchers evaluated the effect of adding another nanomaterial (nano calcium carbonate) and performed dynamic modulus tests.

Zhu et al. [46] presented a study similar to the one reported by [14]. The researchers used a nano-ZnO surface modified with c-(2,3-epoxypropoxy) propyltrimethoxysilane and OEVMt to modify three binders (AC 71, 86, and 105 dmm). Initially, the modified nano-ZnO (1 wt%) + OEVMt (3 wt%) was mixed with the binders at 150 ± 5 °C and 4000 rpm for 60 min using a high shear mixer. Mixing was then continued using a paddle agitator at 2000 rpm for 90 min. The same process was performed for the unmodified binders. The binders were aged in TFOT, PAV, UV, and NEA (the standard TFOT sample was exposed to natural light for 90 days). Rheology tests were performed using DSR and BBR. The modifiers improved the resistance to rutting, thermal oxidation, and photooxidation aging. The enhancement effect of the anti-aging modifier depended on the nature of the binder. The anti-aging modifier improved the low-temperature rheological performance of the AC 90 dmm binder, and had little influence on the others. Zhu et al. [47] published the results of a similar study which evaluated aging resistance according to FTIR testing.

Zhang et al. [12] modified an AC of 64.3 dmm with three ZnO nanoparticles of different spherical sizes (2 microns, 80 nm, and 350 nm average diameter). The ZnO surface was modified using a silane coupling agent. ZnO particles were added in ratios of 2, 3, and

4 wt%. SBS was also added in ratios of 3 and 4wt%. The mixing between AC and ZnO was initially performed at 160 °C, 1500 rpm, and 20 min, and then mixing was continued at 170 °C, 4000 rpm, and 40 min. On the binders, the researchers simulated aging at TFOT. Penetration, softening point, ductility (10 °C), and viscosity tests were performed. Zhang et al. [12] also assessed rheological characterization by means of DSR and BBR tests. The researchers manufactured and designed four asphalt mixtures (AC unmodified; AC modified with 4 wt% ZnO; AC modified with 4 wt% SBS; and AC modified with ZnO and SBS). Marshall, freeze-thaw splitting, rutting, and three-point bending beam tests were performed on these mixtures. The best performance of the evaluated tests was reported as the ZnO size decreased. The results of the study recommended the 80 nm size for an AC modifier.

Rocha Segundo et al. [28] applied three aqueous solutions with pH 8 onto the surface of two asphalt mixtures (named AC 6 and AC 14) to promote their photocatalytic, superhydrophobic, and self-cleaning capabilities. The aqueous solutions contained the following: (1) TiO₂ nanoparticles (4 g/L); (2) ZnO microparticles (1 g/L); (3) TiO₂ nanoparticles; and (4 g/L) + ZnO microparticles (1 g/L). After the spraying of the solution, the physicochemical and morphological properties of the binders' samples were evaluated by AFM, FTIR, and WCA to characterize their hydrophilicity/hydrophobicity. The researchers fabricated mixtures and performed ITS tests on them. Photocatalytic, superhydrophobic, and self-cleaning properties were guaranteed for the AC 14 blend (no deterioration in physical properties) when TiO₂ and TiO₂ + ZnO aqueous solution was used. In the case of the AC 6 mixture, these properties were guaranteed when the TiO₂ + ZnO solution was used. Using the solution containing only ZnO affected the physical integrity of the binder. According to the researchers, pavement surfaces with aqueous solutions of this type could degrade gases such as SO₂ and NO_x; avoid accidents by removing small dirt particles; degrade oils on the pavement surface; and, additionally, prevent pore clogging occurring in porous asphalt mixtures. Benefits for road safety and the environment are, therefore, expected with the use of this technique.

Rocha Segundo et al. [23] modified an AC 38 dmm by combining it with 0.3, 0.8, and 1.0 wt% TiO₂ + 0.2 wt% ZnO (particle size 45 µm). The mixing was performed at 160 °C for 5 min at 200 rpm. FTIR, penetration, softening point, dynamic viscosity (150 °C), and DSR tests were performed on the modified binder. According to the researchers, the modified binder could be used to produce photocatalytic asphalt pavements without physicochemical or rheological impacts on the binder. Both microparticles increase the short-term aging resistance of the binder.

Saltan et al. [48] modified an AC 62.2 dmm (PG 64–22) with ZnO (1, 3, and 5 wt%). Both materials were mixed at 4000 rpm and 160 °C for 2 h. The modified binder was aged in a RTFOT (rolling thin film oven test) and PAV. Its viscosity was measured, and its fatigue and rutting performances were evaluated via DSR and BBR tests. The researchers manufactured HMA and measured moisture susceptibility (ITS) by performing Lottman-modified tests. The best performance of the blends was obtained with the 5 wt% ZnO modification. At 1wt%, the mixture performed similarly to the control sample. The HMA experienced higher resistance against moisture for all modification percentages. ZnO modifications had lower fatigue performance, but the highest rutting and aging performances.

Xu et al. [49] modified an AC 69 dmm with nano-ZnO powder (average particle size 30 nm). The additional percentages were 1, 2, 3, 3, 4, and 5 wt%. The mixing was performed at 150 ± 5 °C at a rotation speed of 5000 rpm for 60 min and stirred with a paddle mixer at a rotation speed of 2000 rpm for 90 min. Penetration, softening point, ductility (5 °C), storage stability, DSR, and LAS tests were performed on the modified binder. Characterization tests were performed on TFOT and UV-aged samples. The nano-ZnO increased the stiffness of the binder and its resistance to UV aging. It was also able to significantly improve its rheological properties. The reasonable dosage of nano-ZnO was quantified at 3 wt%.

Al-Mistarehi et al. [27] modified an AC 68 dmm with Nano Clay—NC (8, 10, and 12 wt%) and nano-ZnO (20–30 nm; 1, 2 and 3 wt%). Using nano-ZnO, the mixing time with

the binder was initially 20 min at a temperature of 160 ± 5 °C and a speed of 2000 rpm. Then, the mixing time increased to 40 min at a mixing temperature of 170 ± 5 °C and 4500 rpm. Penetration, softening point, flash and fire points, RV (135 °C), DSR, and BBR tests were performed on the modified binders. The binder increased its stiffness and viscosity with nano-ZnO, improving its rutting resistance. It also improved its fatigue resistance and low-temperature performance. The optimum nano-ZnO content was 3 wt%. When considering the cost of the modification, the addition of 3 wt% nano-ZnO was cheaper than the addition of 10 wt% NC, and comparable results were achieved.

Fakhri and Shahryari [50] modified an AC 64 dmm with nano-ZnO (0.2, 0.4, and 0.6 wt%; particle diameter 20–60 nm) and nano-reduced graphene oxide (RGO). Mixing was carried out at 160 ± 5 °C and 4000 rpm for 50 min. The modified binders were tested for ductility (15 °C), Saybolt viscosity (135 °C), softening point, penetration, and specific gravity. The researchers manufactured SMA mixtures designed using the Marshall method and performed immersion Marshall, ITS, boiling water, static creep, pull-off adhesion, and SCB (25 °C) tests on them. Both nanomaterials increased the stiffness of the binder and improved the coating and adhesion with the aggregates. Additionally, both nanomaterials improved the performance of the mixes to moisture damage and rutting, and increased the fracture energy promoting resistance against fatigue crack propagation in dry and wet conditions. The best performance was obtained with 0.6 wt% addition of nano-ZnO.

He et al. [51] modified an AC 65 dmm with rodlike nano-ZnO (0.5, 1, 2, 2, 3, and 4 wt%). Mixing was performed at 150 ± 5 °C for 1 h at 5000 rpm, and the samples were aged in TFOT + UV. The researchers performed penetration tests as well as softening point, ductility (5 °C), DSR, FTIR, UV-visible-infrared-absorption, and SEM observations. The rodlike nano-ZnO increased the binder stiffness and resistance to UV aging. The best performance was reported for 3 wt% addition.

De Sousa Neto et al. [52] modified an AC 53 dmm with nano-ZnO (3, 5, 7 wt%). The mixing was performed at 150 ± 5 °C, 2000 rpm for 90 min. The researchers performed rheology tests (PG, MSCR, LAS, master curve), and the modified binders showed improved performance at high temperatures by increasing consistency; viscosity; stiffness; and resistance to aging, rutting, and fatigue. This was especially true at a nano-ZnO content of 7 wt%.

Du et al. [53] modified an AC 66.5 dmm with ZnO and vermiculite (EV) at ratios of 2, 3, 4, 5, 5, and 6 wt%. The ZnO/EV and binder were mixed at 150 °C for 1 h at 3600 rpm, and then mixing was continued using a mechanical stirrer at 2000 rpm for 1.5 h. The asphalts were aged in RTFOT, PAV, and UV. The researchers manufactured asphalt mix and subjected it to STOA. Penetration, softening point, ductility (15 °C), RV (135 °C), DSR, and BBR tests were performed on the modified asphalt binder. Marshall, low-temperature indirect tensile (−6, −12, −18 °C), freeze–thaw splitting, and indirect tensile fatigue (25 °C) tests were performed on the mix. ZnO/EV increased the binder stiffness and rutting resistance at high temperatures, but decreased its ductility and compromised its low-temperature performance. With 5% ZnO/EV, the modified asphalt mix achieved better high- and low-temperature performance, lower susceptibility to moisture, and higher fatigue resistance than the unmodified asphalt mix.

Fu et al. [54] modified an AC 85 dmm with nano-TiO₂, nano-ZnO (nano-TiO₂/ZnO = 2, 4, 6 and 8 wt%), and basalt fiber—BF (6 wt%). The nano-TiO₂ and nano-ZnO were mixed and stirred at a 1:3 ratio for 5 min at 160 °C. Then, both materials were mixed with the asphalt binder for 10 min. Subsequently, shear was applied at 5000 rpm for approximately 50 min. Finally, BF was added and mixed at 2500 rpm for 40 min. The binders were aged at TFOT, PAV, and UV. DSR, ZSV, MSCR, LAS (25 °C), BBR, FTIR tests and observations in SEM were performed on the modified binder. Nano-TiO₂, nano-ZnO, and BF increased rutting, intermediate temperature fatigue, and aging resistance. BF decreased low-temperature fatigue performance, but the addition of nano-TiO₂/ZnO may have helped to increase it. The optimum nano-TiO₂/ZnO dosage was 4wt%.

Kamboozia et al. [55] modified an AC 68 dmm with ZnO (2, 4, 6, and 8 wt%). Both materials were mixed at temperatures between 140 to 155 °C, under 2000 and 5000 rpm, for 60 to 70 min. To disperse the ZnO in the binder, Kamboozia et al. [55] used kerosene as a solvent (it is not clear what the effect of this solvent had on the results). The binders were tested for their softening points, penetration, ductility, and RV (120, 135, 150 °C) to determine the optimum ZnO content (6 wt%). Porous mixtures were manufactured using a SGC and then designed using the Cantabro, Marshall, and ITS tests. Additional Wheel Track Test (45 °C) and SCB (−15, 0, 15 °C) tests were performed on the designed mix. The addition of ZnO increased the binder viscosity, adhesion, and cohesion of the mix. Additionally, it improved the rutting resistance of the porous mix. However, in the SCB test, it was found that the performance of the mixes with ZnO was not the best.

Guo and Zhang [56] modified an AC 64.3 dmm (PG 64-16) with different ZnO morphologies: agglomerate, sphere, rod, and flower. AC was modified with 2, 3, and 4 wt% of ZnO (particle average size was 2 µm). Initially, mixing was performed at 160 °C for 20 min (2000 rpm), and then mixing was conducted at 180 °C for 40 min (4000 rpm). Penetration, softening point, ductility (10 °C), RV (120 °C), DSR, and BBR tests were performed on the modified binders. The researchers manufactured asphalt mixtures designed by the Marshall method and measured the TSR, DS, flexural strength, and flexural tensile strain (three-point bending beam test) on them. ZnO increased the viscosity, stiffness, high-temperature PG, rutting, and aging resistance of the asphalt binder. However, the morphology of the ZnO particle had no effect on the softening point, ductility, penetration, anti-aging ability, or low-temperature ability of the asphalt binder. It also had no effect on the water stability of the asphalt mixture. ZnO particles could improve the DS and water stability of an asphalt mixture. When the morphology of ZnO is rod-like, the properties of asphalt binder and asphalt mixture are better.

Di et al. [57], using nano-ZnO (4 wt%) and Nano-TiO₂ (4 wt%), modified two ACs with two different wax contents, but the physical properties of the ACs are not clear. Researchers show that the rheological properties and wax content of both ACs are their main properties. The modification was performed at 155 °C and 5000 rpm for 30 min using a high shear mixer. LAS, MSCR, and NMR spectroscopy tests have been performed on the modified binders. Both additives improved the aging, rutting, and fatigue resistance of the binder, and nano-ZnO had a greater influence on the fatigue life of the asphalt compared to nano-TiO₂. The antioxidant mechanism of nano-TiO₂ is to inhibit the loss of aromatic carbon, while the oxidation of a nano-ZnO-modified binder causes sufficient aromatization to compensate for the influence of different degrees of aging.

Han et al. [58] initially modified an AC 70 dmm (PG 64-16) with a high content of linear elastomer SBS 791-H (6.5 wt%) by mixing them at 185 °C for 35 min at a speed of 5000 rpm. This SBS-binder was then modified with nano-ZnO, nano-TiO₂, and nano-SiO₂ to attempt to improve its anti-aging performance. The content of all anti-aging agents was 2 wt%. Rheology tests (DSR), FMI, and infrared spectroscopy tests were performed on the modified binders. The nanoparticles did not show significant effects on the rutting, the fatigue, or the aging resistance of the SBS-modified binder. This was mainly due to the weak physical interaction between the nanoparticles and the SBS. Furthermore, no obvious chemical reaction was observed.

Staub de Melo et al. [59] modified an AC 60 dmm (PG 58) with ZnO (3, 6, 9, 12, and 15 wt%). The modification was performed at a temperature of 150 ± 5 °C using a high shear mixture at a speed of 6000 rpm for 15 min. Staub de Melo et al. [59] used a DSR to measure the PG at high temperatures, then used aging index (AI) and non-recoverable creep compliance tests, as well as LAS (20 °C) tests. ZnO increased the stiffness, high-temperature PG, rutting, and aging resistance of the asphalt binder. The fatigue strength increased with ZnO at low strain amplitudes, but decreased at high strain amplitudes.

4. Discussion

The results of the review were presented in Section 3. A summary of this review was demonstrated in Tables 1 and 2. Some aspects to take into account are presented below.

The use of ZnO as a modifier of binders and asphalt mixtures is a recent technique. The first reference included dates from 2015. For this reason, there is currently no technical standard that denotes the minimum conditions which ZnO must meet as a modifier of asphalt binders, nor is there a technical standard that recommends the production of the modification (ZnO content, temperature, mixing time, etc.), the mixing process with the aggregates, the process and/or construction methods in the field, etc.

On the other hand, despite the advances achieved to date in research, there are multiple limitations on its use on an industrial scale in highway pavement construction projects. For example, the uses of this technology in the long term (changes in physicochemical properties, durability, among others) are unclear, and the interactions between the modified binder and the aggregate are not clear either (few studies have been carried out evaluating the performance of asphalt mixes; approximately 33.3% of the consulted documents show tests assessing asphalt mixtures). No study has been found that has evaluated the performance of this technology in full-scale test sections. Only 16% of the consulted studies carried out some type of test under cyclic load in asphalt mixtures. Knowledge about the mechanical performance of asphalt mixes at intermediate, and tests at low temperatures are scarce (approximately 29% of the documents consulted show rheology tests at low temperatures on the modified binder and 16% on asphalt mixtures). As a main limitation of the technology, only slight knowledge in terms of environmental and socio-economic impact can be reported. In short, further research and analyses of life cycle costs should be carried out on this subject in order to recommend this novel technology.

Despite the aforementioned limitations, studies agree that ZnO is a promising material as an asphalt binder modifier, since it tends to improve adhesion with aggregates and resistance to aging (mainly UV aging), rutting, and moisture damage. In addition, it has been shown as a material with low toxicity and low risk to human health that may improve urban air quality.

5. Conclusions

Based on the review carried out herein, it can be concluded:

- (i) ZnO nanoparticles, as binders and asphalt mix modifiers, have been little studied. The first studies date back to 2015.
- (ii) Most studies have evaluated the influence of ZnO on asphalt binder properties. Few studies have been conducted to evaluate the performance in asphalt mixtures.
- (iii) Few studies have evaluated the effect on the environment of modifying asphalt binders with ZnO. The effect of ZnO as a binder modifier in the environment is not clear. Future studies should be carried out in this regard, and studies evaluating socio-economic impacts are also scarce.
- (iv) ZnO contents generally range from 1 to 5% by weight of the asphalt binder. Few studies have used higher contents.
- (v) No studies have evaluated the effect of ZnO as a filler in asphalt mixtures.
- (vi) Most studies have been conducted to evaluate the effect of ZnO on aging resistance. All researchers conclude that ZnO is a material that increases the aging resistance of asphalt binders (especially to UV radiation).
- (vii) ZnO tends to increase the stiffness of the binder, improving its resistance to rutting in high-temperature weather. It also tends to increase adhesion with aggregates, improving resistance to moisture damage. Concerning fatigue, most studies conclude that ZnO increases binder strength. At low service temperatures, the performance of ZnO is not clear, and few studies have assessed this.
- (viii) The effect of ZnO changes depending on the size of the nanoparticles, its morphology, and the type of modified binder.

- (ix) To date, no studies have been found that evaluate the performance of ZnO in full-scale tests. For this reason, its long-term influence on the performance of asphalt mixes is not clear.

For the previously mentioned reasons, future studies should focus on: (i) evaluating the effect of this technology on the environment and conducting economic analyses; (ii) carrying out further research to evaluate the effect on the properties of asphalt mixtures (modified binder-aggregate interaction); (iii) measuring long-term properties; (iv) evaluating the effect on the properties of mixtures at low temperatures; and (v) measuring, in greater depth, the physical–chemical interaction of ZnO with binders and asphalt mixtures.

Finally, for future pavement construction projects, ZnO should be viewed as a promising material from both a technical (as an asphalt binder modifier) and an environmental point of view.

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