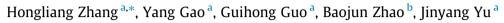
Construction and Building Materials 159 (2018) 578-586

Contents lists available at ScienceDirect

Construction and Building Materials

journal homepage: www.elsevier.com/locate/conbuildmat

Effects of ZnO particle size on properties of asphalt and asphalt mixture



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HIGHLIGHTS

• Three sized ZnO nanoparticles were made.

• The decrease of ZnO size in asphalt can change its properties.

• The smaller ZnO size could change the properties of asphalt mixture.

ARTICLE INFO

Article history: Received 18 May 2017 Received in revised form 2 November 2017 Accepted 7 November 2017

Keywords: Micro and nano ZnO Size Asphalt Asphalt mixture Laboratory performance

ABSTRACT

Micro or nano zinc oxide (ZnO), as a modifier, can significantly improve some properties of asphalt and asphalt mixture, but the effect of ZnO particle size is still unclear. In this paper, three different spherical sized ZnO nanoparticles (2 μ m, 80 nm and 350 nm in average diameter) were made by homogeneous precipitation method and sol-gel method. Asphalt binders and asphalt mixtures modified by ZnO and ZnO/SBS (styrene-butadiene-styrene) were produced by means of a high shear mixer. The effects of ZnO particle size on physical properties of asphalt were analyzed through penetration test, softening point test, ductility test, dynamic shear rheological test (DSR), bending beam rheometer test (BBR) and aging test; and the effects on asphalt mixtures were characterized using the Marshall test, freeze-thaw splitting test, rutting test and a three-point bending beam test. Results show that the decrease of ZnO size in asphalt binder can weaken its creep stiffness, and increase its softening point, ductility, viscosity, anti-rutting factor, creep rate and anti-aging ability. In addition, with the decrease of ZnO size, the high temperature stability, water stability and low temperature crack resistance of asphalt mixture are improved. Finally, based on the test results, the optimum size of ZnO particle is determined as 80 nm.

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1. Introduction

Today researches on nanostructures are very popular because they are being used and pursued for optics, optoelectronics, catalysis, biological sciences and piezoelectricity. Atoms at the surface of particles will have highly activeness when materials are at the nanometer size, and the resulting materials are significantly different from macroscale materials composed of the same substance. Research in the nanostructures of materials has skyrocketed over the last decade.

A variety of micro and nano materials have been used and studied to modify asphalt and concrete engineering, and the effects of nano materials were discussed. Amir et al. [1] found that nano clay and nano hydrated lime (HL) cloud enhance the tensile strength ratio (TSR) of asphalt mixes. Khodary et al. [2] proved that nano-

* Corresponding author. *E-mail address:* zhliang0105@163.com (H. Zhang). size cement bypass could increase the physical, chemical, and rheological properties of modified nanomaterial-asphalt mixture for its high surface area. Khodary et al. [3] analyzed the impact of nano scale CaO on the stiffness properties of asphalt. Liu et al. [4] discovered that the nano CaCO₃ had an advantage influence on the high temperature stability of the asphalt and asphalt mixtures. Tang et al. [5] found that nano-montmorillonite (Nano-MMT)/SBS can improve the high-temperature anti-deformation ability and decrease the softening point of the aged asphalt. Yao et al. [6] explored the effects of nano silica on the rutting and fatigue cracking resistance of asphalt binders. Xiao et al. [7] showed that carbon nanoparticles was helpful in enhancing the failure temperature, complex modulus, and elastic modulus values and in increasing rutting resistance of the RTFO binder. Guo et al. [8] prepared modified nano TiO₂ by co-precipitation method and studied the influence of nano TiO₂ on the properties of coal tar pitch. Zhang et al. [9] made spherical nano Fe₃O₄ particles modified asphalt successfully. In addition, nano-SiO₂ has been reported to enhance concrete







workability and compressive strength [10-12], to improve resistance to water penetration [13], and to contribute to control the leaching of calcium [14]. Nano-TiO₂ could act as a trigger with the photocatalytic degradation of pollutants when it was contained in concrete [15,16]. Nano-Al₂O₃ has been shown to obviously improve the modulus of elasticity of asphalt [17].

However, the micro or nano materials with single size were often used in those researches listed above. Only several scholars had studied the effects of the size of HL on the properties of asphalt or asphalt mixture. Shen et al. [18] reported that sub nano-sized HL can improve about 10% in tensile strength ratio (TSR) for HMA. Aboelkasim et al. [19–21] suggested that the nano sized hydrated lime (NHL) could perform potential benefits compared with regular-sized hydrated lime (RHL) as a modifier of asphalt.

ZnO, non-toxic, odorless, gender-based oxide, is an important semiconductor material. And it has been used in different areas for its special characteristics [22,23]. Recently, some scholars have focused on the effects of ZnO on the properties of asphalt and asphalt mixture. Gholam et al. [24] found that adding nano ZnO could improve the total surface free energy (SFE) of the asphalt binder. Zhang et al. [25] showed that ZnO could change the compatibility between asphalt and polymer and further affect the dispensability of polymer in asphalt binder. Xiao et al. [26] found that ZnO nanoparticles played an effective role in decreasing creep potentials of the asphalt mix samples. Kang et al. [27] found that the nanocoating could improve the tensile strength ratio of the hot mix asphalt. Liu et al. [28] studied the functions principles of the ZnO and SBS modified asphalt. In summary, the recent researches showed that ZnO could significantly improve the properties of asphalt and asphalt mixture. However, the effects of ZnO particle size on properties of asphalt and asphalt mixture are still unclear

The aim of this paper is to study the effects of ZnO particles size on property of asphalt and asphalt mixture. In this paper, the spherical ZnO particles with different size were made in laboratory. Then, the ZnO particles were used to prepare micro and nano ZnO modified asphalt and ZnO/SBS modified asphalt. The effects of ZnO particle size on the properties of asphalt and asphalt mixture were analyzed by a serious of tests.

2. Materials

2.1. Nano ZnO

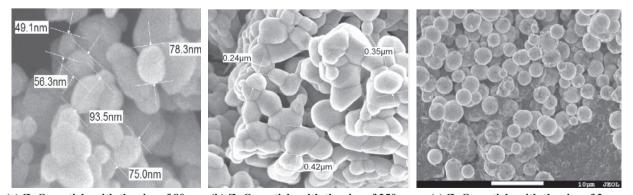
Over the past few years, various methods have been directed towards achieving micro or nano structured ZnO, such as sol-gel method, homogeneous precipitation method, micro emulsion method, combustion synthesis method and hydrothermal method. This paper used homogeneous precipitation method and sol-gel method to synthesize micro or nano structured ZnO, because the reactions in those two methods could occur at room temperature and do not require expensive equipment [29–32].

With zinc acetate, zinc nitrate and absolute alcohol as the starting materials, oxalic acid and urea as precipitating agents, acetyl trimethylammonium bromide as the modifier, the micro and nano ZnO particles were obtained by homogeneous precipitation method and sol-gel method. ZnO particles with average diameter of 80 nm, 350 nm and 2 μ m were got by changing the reaction conditions.

ZnO particle with the size of 80 nm was produced by the following procedures: (1) 6.6 g of zinc acetate was dissolved completely into 50 ml deionized water at 75 °C to get acetate solution, and 0.13 g of acetyl trimethylammonium bromide was dissolved completely into zinc acetate solution; (2) 8 g of oxalic acid was dissolved completely into 150 ml absolute alcohol at 75 °C to get anhydrous ethanol solution with oxalic acid; (3) anhydrous ethanol solution with oxalic acid was slowly distributed into the zinc acetate solution to get the mixed solution; (4) rotors was kept stirring in the mixed solution at 75 °C until the white wet gel was got; (5) the white wet gel was rinsed for two times with deionized water and absolute alcohol and then dried at 80 °C; and (6) the powders obtained after dried were calcinated at 450–550 °C for 3 h. Finally, the nano powder with the average particle size of 80 nm was got (Fig. 1(a)).

ZnO particle with the size of 350 nm was produced as follows: (1) 3.5 g of zinc acetate was dissolved completely into 80 ml deionized water of 45 °C to get zinc acetate solution, and 1.44 g of oxalic acid was dissolved completely into 160 ml deionized water of 45 °C to get oxalic acid solution; (2) oxalic acid solution was slowly distributed into the zinc acetate solution to get anhydrous ethanol solution with oxalic acid; (3) the rinsed and dried steps were same with those when the ZnO with the size of 80 nm was produced; and (4) the powders obtained after dried were calcinated at 600 °C for 4 h. Finally, the nano powder with the average particle size of 350 nm was got (Fig. 1(b)).

ZnO particle with the size of 2 μ m was produced by the following steps: (1) 30.7 g of zinc nitrate was dissolved completely into 200 ml deionized water to get zinc acetate solution, and 12 g of urea was dissolved completely into 200 ml deionized water to get urea solution; (2) the two solutions were poured into a sealed container and were stirred continuously at 80 °C for 2–3 h; (3) the rinsed and dried steps were same with those when the ZnO with the size of 80 nm was produced; and (4) the powders obtained after dried were calcinated at 600 °C for 4 h. Finally, the powder with the average particle size of 2um was got (Fig. 1(c)).



(a) ZnO particle with the size of 80nm (b) ZnO particle with the size of 350nm

n (c) ZnO particle with the size of 2um

Fig. 1. Spherical ZnO particles with different sizes.

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