

# Effect of cement on the demulsifying behavior of over-stabilized asphalt emulsion during mixing



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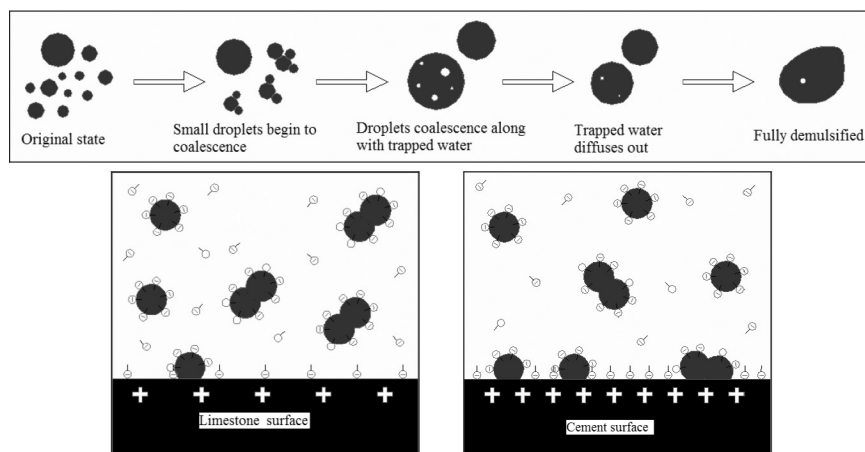
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## HIGHLIGHTS

- Effect of pH value and  $\text{Ca}^{2+}$  on the rheological properties of asphalt emulsion are studied.
- The mixing stability of asphalt emulsion with cement and limestone is studied.
- The demulsifying behavior of asphalt emulsion with cement and limestone is discussed.
- The direct asphalt droplet adhesion on cement is dominated during mixing.

## GRAPHICAL ABSTRACT



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## ABSTRACT

Over-stabilized asphalt emulsion is normally used in cement asphalt emulsion (CA) composites. The mixing stability of asphalt emulsion with cement is vital to the workability and mechanical properties of CA composites. However, the effect of cement on the demulsifying behavior of asphalt emulsion during mixing is not clearly understood. To reveal the demulsifying behavior, effect of pH value and  $\text{Ca}^{2+}$  on the rheological properties and mixing stability of over-stabilized asphalt emulsion are studied. Results indicate that cationic emulsions have no obvious change in the tested pH value and  $\text{Ca}^{2+}$  concentration range.  $\text{Ca}^{2+}$  ion is very harmful to the chemical stability of anionic emulsion. There are three stages in the demulsifying process of pure emulsion: (1) small asphalt droplet coalescence along with trapped water; (2) trapped water diffuses out from coalesced asphalt droplets; (3) complete demulsification. There are two demulsifying behaviors when asphalt emulsion is mixed with fine solid. One is asphalt droplet coalescence, which is dominant in the mixing with limestone powder. The other is the direct droplet adhesion on solid surface, which is dominated in the mixing with cement. This difference is due to that the adsorption ability of cement is much stronger than that of limestone powder. The ions of  $\text{OH}^-$  and  $\text{Ca}^{2+}$  released by cement hydration cannot greatly destabilize asphalt emulsion. However, emulsions with better resistance to the invasion of  $\text{OH}^-$  and  $\text{Ca}^{2+}$  still have a better mixing stability. Therefore, reducing the adsorption ability of cement and increasing the resistance of emulsions to hydrate ions can improve the mixing stability of asphalt emulsion with cement.

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## 1. Introduction

Cement asphalt emulsion (CA) composites are very promising materials suitable for many applications. In the road industry, cold mix asphalt and cold recycling asphalt mixtures are more preferred than hot mix asphalt in pavement maintenance and repair due to the low energy consumption and atmospheric emission of asphalt emulsion [1–3]. In the ballastless slab track of high-speed railway, grouting cement asphalt emulsion mortar (hereinafter abbreviated CA mortar) has been greatly appreciated as the elastic cushion and even-adjusting layer between slab track and concrete base because of its good damping effect and high workability [4,5]. In all these composite materials, asphalt and cement are both considered as binders, thus these composite materials can be combined with the merits of cement (high strength) and asphalt (good flexibility) to be good semi-flexible materials [6–8]. The temperature sensitivity of CA composites can be greatly improved compared to asphalt mixture, and the ductility becomes much better than pure cement-based materials [6–8]. Besides, asphalt emulsion is a low-viscosity aqueous emulsion at room temperature. It can be mixed well with other water-based binder like cement [9]. This property ensures that CA composites have a good workability at room temperature.

Theoretically, CA composites are more advantageous than cement-based materials and asphalt-based materials. However, there are still scores of challenges in the application of CA composites. A crucial issue in CA systems is the mixing stability of asphalt emulsion. Asphalt emulsion is inherently unstable. It is easily demulsified in or after mixing with other materials, especially for cement due to its fine size and high reactivity. The poor mixing stability of asphalt emulsion with cement not only degrades the constructability of CA composites, but also affects the mechanical properties of CA composites. For grouting CA mortar, the high flowability may be suddenly lost during mixing due to the inadequate mixing stability of asphalt emulsion [10]. For cold mix asphalt and cold recycling asphalt mixture, aggregate coating is vital to their mechanical properties [11]. Due to inadequate mixing stability, asphalt emulsion may quickly separate from water during mixing and cannot fully coat aggregates [11]. Besides, the quickly demulsified asphalt does not adhere to aggregate because of a water-rich phase between demulsified asphalt film and aggregate, and the trapped water in asphalt is more difficult to squeeze out than free water [12]. As a result, poor mechanical properties are obtained.

Even though asphalt emulsion has not strong demulsifying behavior during mixing, asphalt emulsion with good mixing stability is still beneficial to the workability and mechanical properties of CA composites [10]. Improvement in the mixing stability of asphalt emulsion can reduce the water to cement ratio of CA paste at the same viscosity, which may be beneficial to the mechanical properties of hardened CA paste [10]. Improvement in the mixing stability of asphalt emulsion reduces the initial viscosity of CA paste [10,13], which can retard the demulsification process of asphalt emulsion and prolong the workable time of CA paste [13]. That is why over-stabilized asphalt emulsions with high dosage of emulsifier are generally recommended for CA mortar [14,15]. Nowadays, over-stabilized asphalt emulsions with high dosage of emulsifier are also preferred in cold mix asphalt and cold recycling asphalt mixtures in China [16].

Extensive studies have been performed on the interaction between asphalt emulsion and cement in CA paste at fresh state in recent years. On the one hand, asphalt emulsion has a strong retarding effect on cement hydration that is related to emulsion type, emulsion dosage, and emulsifier type [17–19]. On the other hand, cement hydration, which changes the properties of a CA system (e.g., pH value and ion concentration) [20], induces

demulsification and accelerates the demulsifying rate. Thus, the variation of the fresh properties (e.g. viscosity and flowability) of CA paste is mainly related to the demulsification of asphalt emulsion [13]. The adsorption behavior between cement particles and asphalt droplets, the variations of droplets morphology and droplets size distribution are three main physical changes of asphalt droplets in demulsification, which all affect the viscosity of CA paste [13,21]. Therefore, viscosity can be used as an index of the demulsifying behavior in CA paste [10,13,21–23]. In fact, viscosity is usually used as an index of the physical properties of asphalt emulsion [24–27].

The function of emulsifier is to emulsify asphalt and prevent coalescence of asphalt droplets once formed. Therefore, the mixing stability of asphalt emulsion with cement is controlled by emulsifier [28,29] and in particular by the interaction between cement and emulsifier. There are three main effects when cement is added into asphalt emulsion. The first one is the change of pH value, which may be great harmful to the stability of most cationic emulsifiers [9,30]. The second one is hydrate ions, mainly  $\text{Ca}^{2+}$ , can interact with emulsifiers and induce precipitation [20,22]. Previous studies indicate that  $\text{Ca}^{2+}$  reduces the Zeta potential of anionic emulsion [20] and makes anionic emulsifier insoluble in aqueous solution [20,22,31]. The last phenomenon of concern is the adsorption behavior between cement and emulsifier when cement is added to asphalt emulsion. Emulsifier is essentially a surfactant whose molecular structure is similar to superplasticizer. Both anionic and cationic emulsifiers can be adsorbed by cement and change the rheological properties of cement paste [14,15,32,33], which can destabilize asphalt emulsion [21].

Although the mechanisms and factors influencing the mixing stability of asphalt emulsion are known when cement is added, there are still many issues that are unclear. Firstly, the previous emulsions used in the analysis on the interaction between asphalt emulsion and cement were usually traditional slow-setting emulsions in which emulsifier dosage is less than 2% [9,34]. However, over-stabilized asphalt emulsions with high emulsifier dosage are now more preferred not only in CA mortar, but also in cold mix asphalt and cold recycling asphalt mixtures in China [16]. Due to high emulsifier dosage and special chemical compositions of emulsifier, over-stabilized asphalt emulsions have significantly different demulsifying behavior compared to traditional slow-setting emulsions. Secondly, most previous studies were based on one charge type of asphalt emulsion, and believed that anionic emulsion has better mixing stability than cationic emulsion [22,31] due to the aqueous alkaline environment caused by cement hydration. However, both cationic and anionic asphalt emulsion are utilized in different types of CA composite, and the mixing stability is not fully dependent on emulsifier charge type. Thirdly, and also the most important, the demulsifying behavior of asphalt emulsion in the mixing with cement is unclear. There are two mechanisms proposed to explain the demulsifying behavior when asphalt emulsion is mixed a solid [35,36]. The first one concerns the emulsifier adsorption on the solid surface followed by the droplet coalescence produced by the reduction of the overall electric repulsion and further adhesion on the solid surface (Fig. 1(a)) [35,36]. However, if the desorption kinetics of the emulsifier from the interface is a slow process compared with the emulsifier adsorption kinetics onto the solid surface, the direct droplet adhesion on the solid surface could be the predominant mechanism, and then droplets are coalesced (Fig. 1(b)) [35]. The first mechanism is usually used to explain the demulsifying process of asphalt emulsion mixed with a solid [9,13]. However, some researchers suggested that the demulsifying process of asphalt emulsion in CA paste was essentially the direct droplet adhesion on cement surface [21]. The demulsifying behavior of asphalt emulsion in the mixing with cement should be further studied.

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