



# Enhancing sustainability in pavement Engineering: A-state-of-the-art review of cement asphalt emulsion mixtures

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## ABSTRACT

Cement asphalt emulsion mixture (CAEM) is an environmentally sustainable substitute for hot mix asphalt and can trigger a substantial economic benefit. This paper systematically reviews the interactions between the organic–inorganic composites and their influence on the performance of CAEM. First, the interactions between asphalt emulsion (AE) and cement are introduced. Next, the demulsification of AE and hydration of cement in the CAEM system are analyzed. Finally, the fresh properties of CA paste, the static and dynamic mechanical properties of CA mortar and its applications in ballastless slab tracks, and the road performance of CAEM and its applications in pavement construction are discussed. This review allows for a better understanding of the interaction of the organic–inorganic composite and thus has a better strategy to regulate the performance of CAEM and promote its practical application.

## Introduction

Cement and asphalt are commonly employed in infrastructure construction, including pavements, tunnels, and bridges (Castillo et al., 2015; Cui et al., 2023b; Liu et al., 2021; Lu and Zhong, 2022). Cement-based materials offer benefits such as high compressive strength and corrosion resistance (Cui et al., 2023a; Huo et al., 2023b; Jiang et al., 2023; Lu et al., 2022c; Lu, Dong et al., 2023c). However, they tend to be rigid and lack deformability (Huo et al., 2023a; Huo et al., 2023b; Lu et al., 2023a; Lu et al., 2023b; Lu et al., 2023). On the other hand, asphalt composites exhibit excellent flexibility, outstanding visco-elastic behaviour, and damping property (Liu et al., 2022a; Rafiq Kakar et al., 2022; Ren et al., 2022). Nevertheless, they are prone to permanent deformation when subjected to repeated loading (Gong et al., 2023; Hong et al., 2021; Liu et al., 2021; Liu et al., 2022a), as well as high energy composition and huge carbon footprint (Lu et al., 2021; Zhao et al., 2023). A notable example of an organic–inorganic composite is the cement asphalt emulsion mixture (CAEM) (Al-Khateeb and Al-Akhras, 2011; Ayar, 2018; Boltryk and Małaszkiwicz, 2013). CAEM possesses a diverse range of properties due to its composition, which primarily includes cement, aggregates (sand), asphalt emulsion (AE), and various admixtures (Buczyński, 2016; Du, 2016, 2018). The

interactions within CAEM involve the effect of cement on the demulsification process of AE and the impact of AE on the hydration of cement (Wang et al., 2014; Xiao et al., 2019; Xu et al., 2017). Moisture released during the demulsification of AE can be utilized for cement hydration (Yan et al., 2017; Yan et al., 2014; Yang et al., 2021). This water helps resolve the conflict between the “water-repellent” nature of AE demulsification and the “water demand” for cement hydration (Zhu et al., 2019a; Zhu et al., 2019b; Zhu et al., 2018). Unlike cement-based or asphalt-based materials, CAEMs not only combine the benefits of both but also compensate for their respective disadvantages (Wang et al., 2020; Wang et al., 2015; Wang et al., 2014). CAEM finds widespread applications, such as polymer-modified concretes, cold recycling of asphalt pavement, back-filling grouts for shield tunnels, semi-flexible base layers, cushion layers for ballastless track systems, and more (Tan et al., 2013; Thanaya et al., 2014; Tian et al., 2017; Tian et al., 2020; Wang et al., 2018).

Current research efforts are focused on conducting various experiments to explore the characteristics of CAEM and investigate the factors that influence its performance (Saride et al., 2016; Shanbara et al., 2018; Shirzad et al., 2018; Tabib et al., 2018). These factors include workability, mechanical properties, and durability (Nassar et al., 2016; Pettinari et al., 2014; Saadoon et al., 2018). The performance of CAEM,

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both in its fresh and hardened states, is intricately linked to the interaction between cement particles and AE (Martínez-Echevarría et al., 2012; Miljković et al., 2019; Moghadas Nejad et al., 2017). The hydration of cement, demulsification of AE, subsequent rheological behavior, and formation of the microstructure of CAEM are all highly dependent on this interaction process (Liu and Liang, 2017; Liu et al., 2014; Liu et al., 2022b). Therefore, a comprehensive understanding of the interaction between cement particles and AE is vital for optimizing the performance of CAEM and facilitating its broader application in engineering structures (Li et al., 2019; Lin et al., 2017; Lin et al., 2018; Lin et al., 2015; Ling et al., 2016).

Fresh CAEM is a mixture formed by dispersing cement particles and asphalt droplets, which are surrounded by emulsifiers, in an aqueous phase (Chen et al., 2020; Dhandapani and Mullapudi, 2023). The cement in CAEM is not only reactive grain that releases hydration ions (such as  $\text{Na}^+$ ,  $\text{K}^+$ ,  $\text{Ca}^{2+}$ ,  $\text{SO}_4^{2-}$ ,  $\text{HSiO}_4^-$ , and  $\text{OH}^-$ ) into the suspension system at different concentrations but also possesses a high adsorption capacity for AE (Dołycki and Jaskuła, 2019; Fang et al., 2022; Guo et al., 2022a). AE, on the other hand, is a thermodynamic metastable system, and its stability is greatly influenced by various parameters such as water loss, pH changes, counter ions, swelling of asphalt droplets, emulsifier dosage, and the admixed solid particles (Guo et al., 2022b; Jain and Singh, 2021; Jiang et al., 2021). The stability of AE is inevitably affected by cement hydration, particularly through the interaction between cement and AE (Guo et al., 2022a; Li et al., 2022; Li et al., 2018). Conversely, the hydration of cement is hindered by the adsorption and subsequent demulsification of AE (Li et al., 2020; Ma et al., 2023). Consequently, the early properties of CAEM are indirectly affected. A precise understanding and evaluation of the interaction between different types of AE and cement are crucial for enhancing the application of CAEM composites in specialized areas like rapid repairs and high-corrosion resistance structures (Ouyang et al., 2016b; Ouyang et al., 2018; Ouyang et al., 2017).

In recent years, there have been extensive investigations focusing on the rheological properties, mechanical properties, and durability of CAEM (Jain and Singh, 2021; Ouyang and Tan, 2015; Ouyang et al., 2016d; Peng et al., 2015; Shanbara et al., 2021). For instance, Zhang et al. (Zhang et al., 2012) conducted a study on the rheological properties of fresh CAEM, examining the influences of AE type, temperature, and curing time. Wang et al. (Wang et al., 2014) investigated the effects of two types of AE (anionic and cationic) on the performance of low-modulus CEAM and high-modulus CEAM, with varying asphalt-to-cement ratios (A/C). Jiang et al. (Jiang et al., 2021) reviewed previous studies on CEAM and summarized the factors influencing its performance. Leiben et al. (Leiben et al., 2018) investigated the damping characteristics of CEAM under various loading frequencies, contributing to understanding its resistance to vibration and deformation caused by repeated loads in engineering structures. Ouyang et al. (Ouyang et al., 2016b) studied the thixotropic behavior of fresh CAEM. These reports have examined different parameters such as the A/C ratio, sand/cement (S/C) ratio, water/cement (W/C) ratio, temperature, and additional additives to evaluate the properties of CEAM (Ouyang et al., 2018; Ouyang et al., 2017; Ouyang and Tan, 2015; Yang et al., 2021; Zeng et al., 2021), there is still a lack of a comprehensive review on the internal interaction between cement and AE. Furthermore, the effect of this interaction on the performance of CAEM remains ambiguous.

To address this research gap, the present review aims to provide valuable insights into the existing controversies by conducting a systematic review and analysis. The primary objective is to emphasize the importance of this study. In essence, this paper offers a comprehensive review of the interaction between cement and AE. It delves into the interaction mechanism and explores its influence on various aspects, including the demulsification of AE, hydration of cement, rheological properties, and microstructural evolution of cement-asphalt emulsion (CA) paste. Additionally, the review provides an outlook and proposes perspectives for further research in this field.

## Interactions between asphalt emulsion and cement grains

Different mechanisms have been reported in the existing reports to elucidate the intricate interactions between AE and cement, considering both physical and chemical perspectives (Li et al., 2018; Li et al., 2020; Ouyang et al., 2017). However, further investigation is required to clearly understand the interaction mechanisms within CA paste before delving into the demulsification of AE and cement hydration. Generally, the interactions between cement and AE include two processes: the adsorption/destabilization of AE and the hydration of cement (Dołycki and Jaskuła, 2019; Fang et al., 2022).

### Adsorption of AE on the cement surface

#### Electrostatic attraction

The electrostatic attraction theory argues that the interaction between AE and cement is primarily a physical effect, as no new crystalline phases have been detected in the CA composite (Li et al., 2020; Ouyang et al., 2016b). The emulsifiers typically used in AE can carry a positive or negative charge, while the hydrated cement grains develop negatively charged silicate hydrates (or positively charged aluminate) on their surface (Tian et al., 2020; Zarei et al., 2022). These charged components provide sites for the adsorption of AE, as illustrated in Fig. 1a. Cationic AE, tends to adsorb on the surface of negatively charged silicate phases, such as  $\text{C}_2\text{S}$  and  $\text{C}_3\text{S}$ . Conversely, anionic AE is primarily adsorbed onto the positively charged  $\text{C}_3\text{A}$  surface (Fang et al., 2022). Furthermore, negatively charged AE can be adsorbed through the complexation between  $\text{Ca}^{2+}$  and  $\text{H}_2\text{SiO}_4^-$  on the silicate phase surface, creating positively charged sites (Guo et al., 2022a). Confirmation of the adsorption of AE droplets on the cement surface can be achieved through the observation of Environmental Scanning Electron Microscopy (ESEM). In fact, Fig. 1b demonstrates that both anionic and cationic AE droplets can adsorb onto the surface of cement particles (Fang et al., 2022).

#### Chemical bonding

Chemical bonding is a theory proposing the occurrence of a chemical reaction between AE and cement (Fang et al., 2022; Li et al., 2018; Li et al., 2020). This theory indicates that the carboxylic acids present in AE can undergo a reaction with alkali groups in the cement, e.g.,  $\text{Ca}(\text{OH})_2$ , leading to the formation of calcium salts (Li et al., 2022; Li et al., 2018). The high chemical reactivity of  $\text{Ca}(\text{OH})_2$  facilitates the creation of ample binding sites for AE within the cement hydrates (Guo et al., 2022b; Jiang et al., 2021). Fig. 2 supports the likelihood of chemical adsorption behavior, as carboxylic acids are primarily found in asphaltenes and resins.

The mechanisms underlying the interaction between cement and AE have not achieved universal acceptance. It is suggested that both the electrostatic adsorption theory and the chemical bonding theory coexist in CA paste. It is important to note that the reaction between cement and AE is highly dependent on time, and further comprehensive investigations are required to understand the mechanism of their interaction across different time scales. Future studies should delve into the temporal aspects and the respective dominance of these theories to gain a deeper understanding of the interaction between cement and AE.

### Destabilization of AE by cement hydration ions

The stability of AE is primarily governed by the emulsifier added to the mixture, which disperses around the AE droplets (Fang et al., 2022; Jiang et al., 2021; Ouyang et al., 2016a). This dispersion leads to the adsorption of ions and counter-ions onto the AE droplets, resulting in the formation of an electrical double layer. This double layer gives rise to a positive or negative charge on the AE droplets (Jiang et al., 2021; Li et al., 2022). By generating a robust double layer, an adequate repulsive force is established, effectively preventing the coalescence of AE droplets and ensuring their stability.

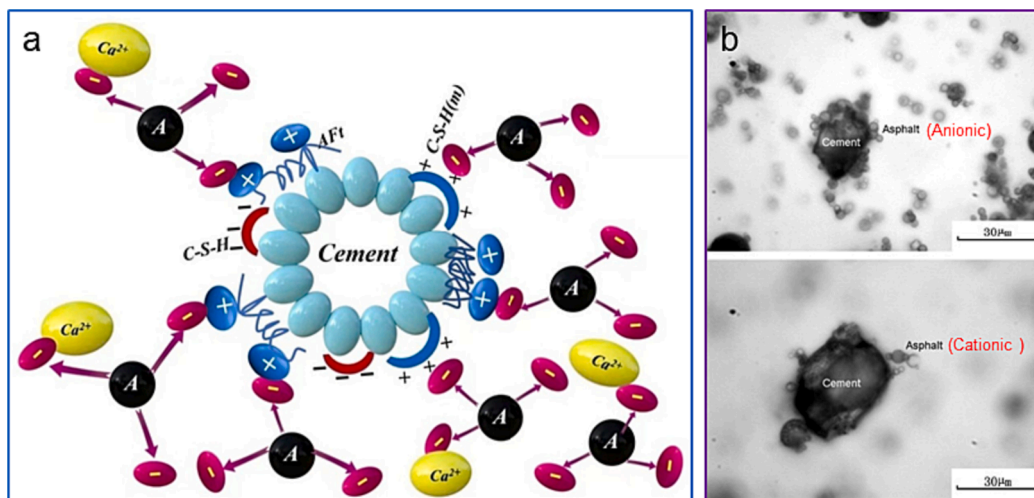


Fig. 1. (a) The electrostatic attraction between AE and cement (Li et al., 2018); and (b) images of CA paste at 5 min (Fang et al., 2022).

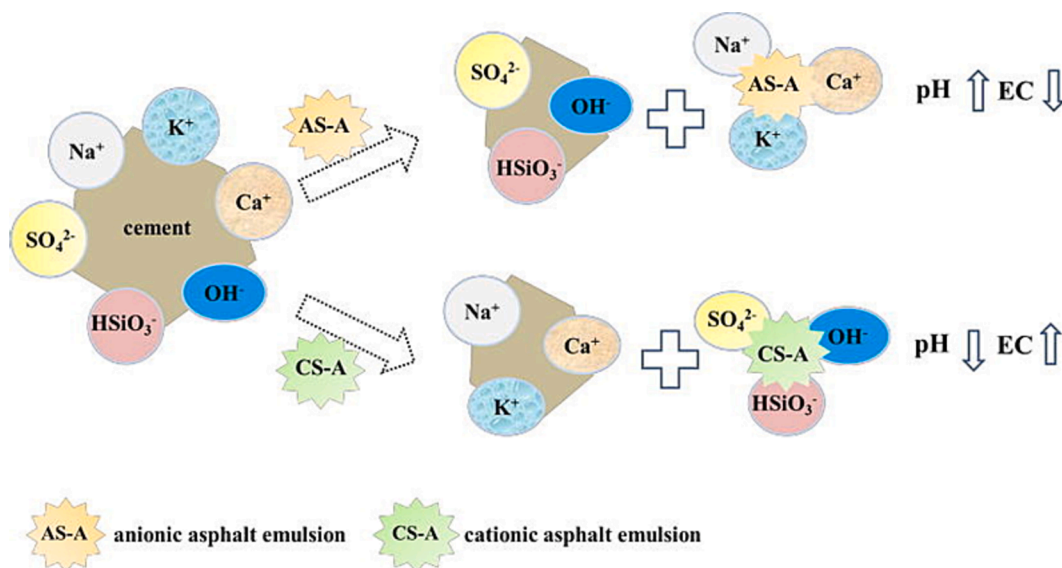


Fig. 2. Schematic of the interaction between two types of AE and cement (Sun et al., 2019).

The changes in the bilayer structure within suspensions can be assessed through the measurement of Zeta potential, which provides insight into the stability of AE (Erickson and Li, 2001; Fang et al., 2022). Variations in pH caused by cement hydration, water loss during cement hydration, and interaction with  $\text{Ca}^{2+}$  ions can alter the bilayer structure on the surface of AE droplets, leading to electrostatic repulsion and influencing AE stability (Li et al., 2018; Li et al., 2020; Ouyang et al., 2016b; Peng et al., 2015). In particular, the precipitation of anionic AE tends to occur at a high concentration of  $\text{Ca}^{2+}$  (Li et al., 2022; Li et al., 2018; Li et al., 2020). As depicted in Fig. 3,  $\text{Ca}^{2+}$  ions released from dissolved cement clinker can interact with emulsifier molecules, resulting in their precipitation and subsequent reduction in the emulsifier concentration surrounding the AE droplets (Fang, X. et al., 2016; Tan et al., 2014; Zarei et al., 2022). This disturbance of the electric double layer can reduce the repulsive force and make the droplets prone to flocculation or coalescence (Fang et al., 2022; Zarei et al., 2022).

The interaction between  $\text{Ca}^{2+}$  and AE can be further elucidated by the observation that introducing cement and  $\text{CaCl}_2$  induces a noticeable colour change in the filtrate. This colour change becomes more prominent with increasing amounts of cement and  $\text{CaCl}_2$  (Fang, Xing et al., 2016; Ouyang et al., 2016a; Ouyang et al., 2016c). While adding

limestone filler does not lead to any colour changes. These findings suggest that adding cement and  $\text{CaCl}_2$  can lead to the precipitation of emulsifiers and result in a decrease in their concentration in the solution. This decrease in concentration contributes to the instability of AE, thereby affecting its performance (Ma et al., 2023; Ouyang et al., 2016b).

In conclusion, the complexation of  $\text{Ca}^{2+}$  ions results in a higher adsorption capacity of cement towards anionic AE compared to cationic AE (Ouyang et al., 2018; Ouyang et al., 2017; Ouyang and Tan, 2015). Physical interactions, including electrostatic attraction, pH changes, and water loss, play a significant role in this interaction (Ouyang et al., 2016d; Peng et al., 2015). However, it is important to consider chemical interactions, particularly with anionic AE, when mixing cement with AE that contains a higher concentration of carboxylic acid components that can precipitate.

#### Demulsification of AE and hydration of cement in the CA system

The contact between AE and cement particles encompasses two interconnected reaction processes, these processes mutually influence and enhance each other (Fang, Xing et al., 2016; Fang, X. et al., 2016;



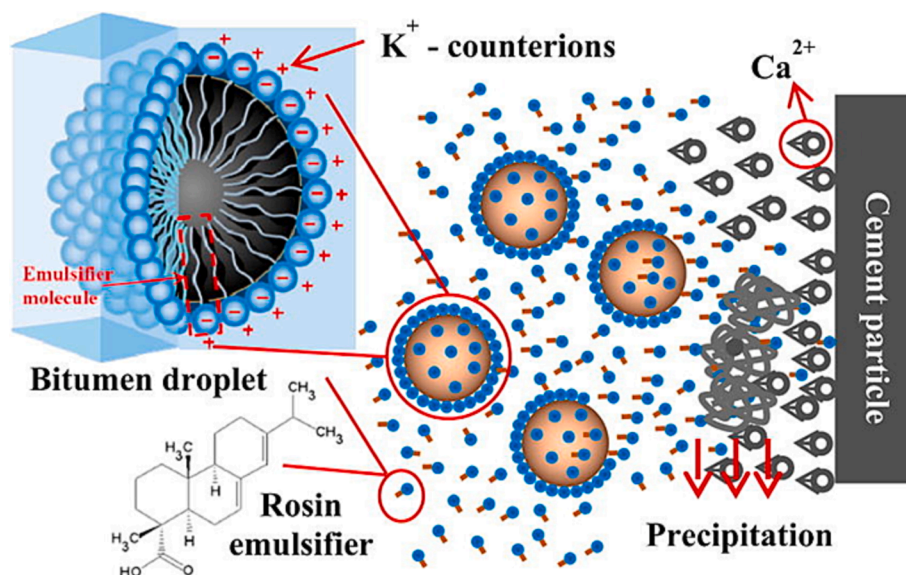


Fig. 3. Illustration of precipitation of anionic emulsifiers in AE in CA paste (Fang, X. et al., 2016).

Ouyang et al., 2016a; Zhang et al., 2012). The water released during the demulsification of the AE can be utilized for cement hydration, while the water consumed during cement hydration facilitates AE demulsification (Tan et al., 2014; Tian et al., 2020). This section explores the processes and mechanisms of both reactions, to gain a deeper understanding of the interaction between AE and cement.

#### Demulsification of AE

The demulsification process of AE is inherently impacted by the interaction between cement particles and AE described earlier. The interaction between cement and AE promotes the cohesion and agglomeration of asphalt droplets, facilitating the development of a continuous asphalt film on its surface (Fang et al., 2022; Guo et al., 2022a; Li et al., 2020). The combined effects of adsorption and instability significantly influence the AE demulsification and the subsequent formation of an asphalt film.

#### Demulsification mechanisms

When AE is mixed with cement particles, the demulsification mechanisms can be categorized into two distinct processes (Fang, X. et al., 2016; Mercado et al., 2012). The first category involves droplet coalescence triggered by the inherent instability of AE, followed by the adhesion of the coalesced droplets onto the solid surface. In the second category, the sequence is reversed, as illustrated in Fig. 4a. Both of these steps play an important role in the demulsification of AE. However, in the mixing process, direct adhesion of droplets onto the cement surface predominates (Ouyang et al., 2016a; Ouyang et al., 2016c). As cement hydrates continue to generate and the AE is broken, the asphalt film gradually envelops the entire cement particle through the accumulation (Zhang et al., 2012), as presented in Fig. 4b.

Typically, the particle size of AE undergoes a significant increase as the mixing time extends (Fang et al., 2022; Fang, Xing et al., 2016). Particles smaller than 5  $\mu\text{m}$  gradually diminish, while the aggregation of particles  $>10 \mu\text{m}$  intensifies. The influence of cation concentration and cation valency on particle distribution becomes more pronounced, particularly in terms of the destabilization of anionic AE (Guo et al., 2022a; Li et al., 2022). In diluted AE, the majority of asphalt exists as individual droplets or very small flocculated droplets.

#### Stability of AE

From a macroscopic perspective, the rheological properties serve as

direct indicators of the stability of AE, with AE instability resulting in an increase in viscosity (Fang, Xing et al., 2016; Ouyang et al., 2016c). The changes in AE viscosity over time are illustrated in Fig. 5. Adding cement can result in a significant improvement in AE viscosity. Particularly, the viscosity experiences a sharp rise when the cement content exceeds 6% (Fang, Xing et al., 2016).

According to a previous study (Fang et al., 2022), the residue content of AE exhibits a gradual increase as the  $\text{Ca}^{2+}$  concentration rises from 0 to 0.1 mol/L. However, when the concentration of  $\text{Ca}^{2+}$  reaches 1 mol/L, the residue content of AE, particularly anionic AE, experiences a sharp increase (Fang et al., 2022; Guo et al., 2022a). This observation suggests that anionic emulsifiers have a higher propensity to precipitate at higher  $\text{Ca}^{2+}$  concentrations (Fang, Xing et al., 2016; Ouyang et al., 2018). This finding aligns with the analysis of anionic emulsifiers and  $\text{Ca}^{2+}$  precipitation and reinforces the significant effect of  $\text{Ca}^{2+}$  on the AE's stability.

#### Hydration of cement

The hardening process of the CA system is more than just a combination of conventional cement hydration and AE (Chen et al., 2020; Dołżycki and Jaskuła, 2019). As previously discussed, the interaction between cement and AE, as well as the subsequent breaking of AE, unavoidably influences cement hydration (Dołżycki and Jaskuła, 2019; Fang et al., 2022). To assess the impact of this interaction on cement hydration, measurements such as heat of hydration, ion concentration, and resistivity of the cement can be employed. The following discussion delves into these aspects.

#### Retardation mechanisms of cement hydration in the CA system

The retarding mechanisms of cement hydration by AE can be divided into two primary aspects (Guo et al., 2022a; Li et al., 2018; Ma et al., 2023; Ouyang et al., 2018): 1) the adsorption of AE on the cement surface (Fig. 6a); and 2) the film layer formed by the emulsification of AE (Fig. 6b).

#### Hydration heat of cement in CA system

The cement hydration in the CA system follows a similar pattern to that of normal cement paste, similar to plain cement hydration (Lu et al., 2022b, c, e). The generation of cement hydrates such as Aft, CH, and C-S-H gels during this process generates heat, indicating the exothermic nature of cement hydration (Lu, Dong et al., 2023b; Lu, D. et al., 2023; Lu et al., 2022a). Notably, the presence of admixed AE significantly



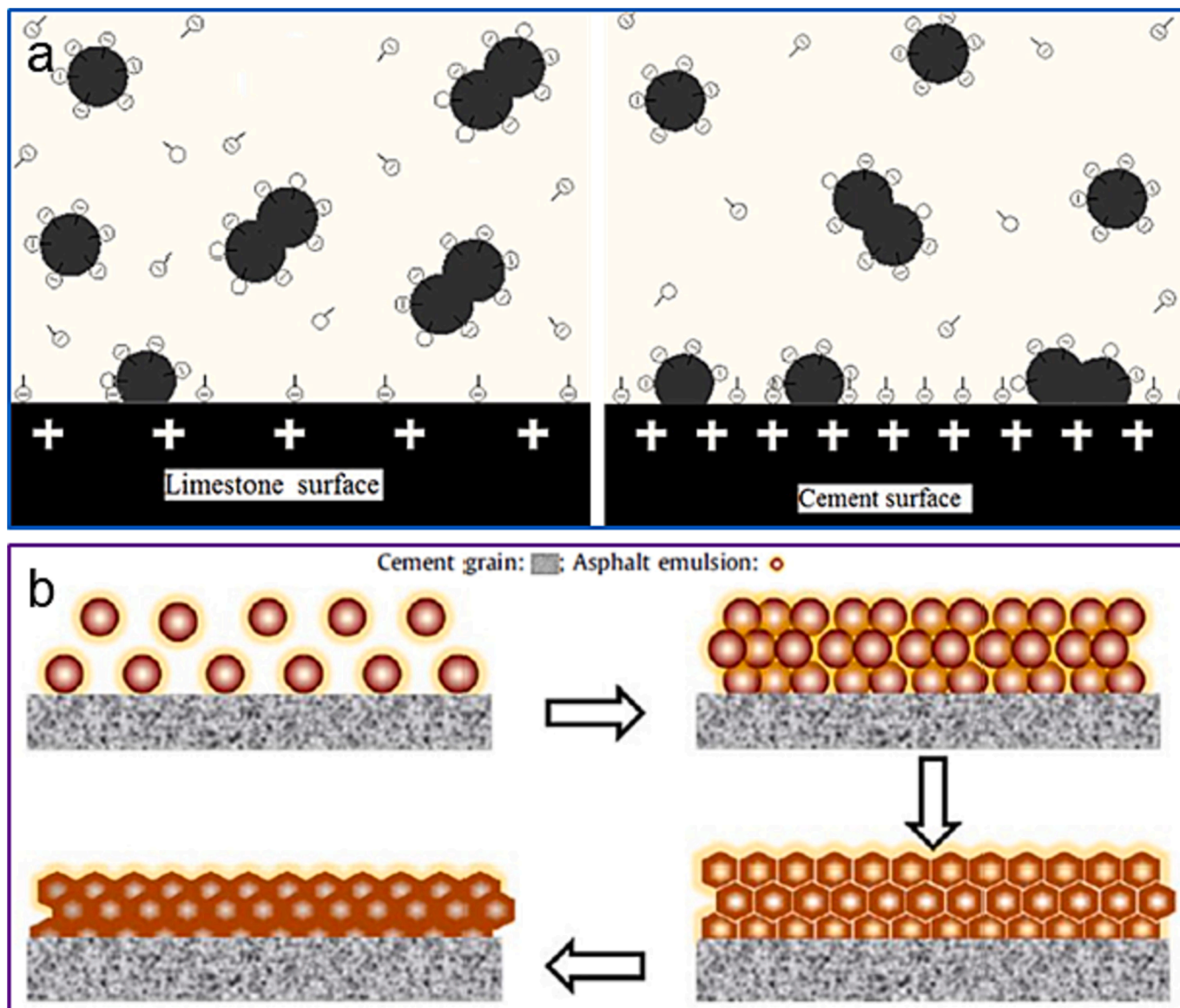


Fig. 4. (a) Demulsification mechanisms of AE (Ouyang et al., 2018); and (b) coagulation and film formation of AE with the presence of cement (Zhang et al., 2012).

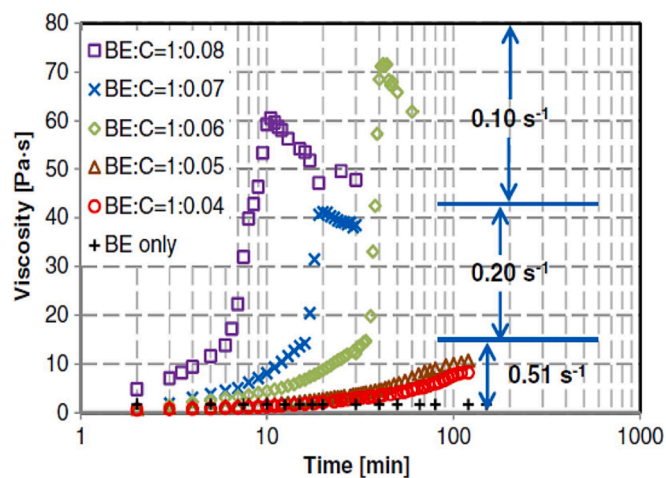


Fig. 5. Viscosity of AE mixture with different dosages of cement. Note, C is the weight ratio of AE and cement (Fang, Xing et al., 2016).

delays the early stages of cement hydration. Higher asphalt-to-cement (A/C) ratios result in longer induction periods, lower total heat of hydration, and slower hydration of CA pastes (Fang et al., 2022; Li et al., 2018). These findings highlight the retarding effect of AE on cement hydration. Specifically, anionic AE exhibits a more pronounced retarding effect compared to cationic AE, primarily due to its strong adsorption onto hydrated cement particles. This phenomenon is depicted in Fig. 7.

#### Ion concentrations of cement in the CA system

The evolution of cement hydration in CA paste is often monitored by analyzing the ion concentrations in the pore solution (Fang, Xing et al., 2016; Fang, X. et al., 2016; Jiang et al., 2020b). It has been observed that the presence of emulsifiers in cement paste results in lower concentrations of  $\text{Na}^+$ ,  $\text{K}^+$ , and  $\text{SO}_4^{2-}$  ions compared to pure cement at the same dissolution period (Fang et al., 2022). This reduction in ion concentrations suggests that the absorbed emulsifiers have a delaying effect on the dissolution process of cement composites (Guo et al., 2022a; Li et al., 2022). Furthermore, the counter ions present in the emulsifiers may react with the anionic groups, forming complex compounds (Li et al., 2018; Li et al., 2020). This reaction can lead to a reduction in the alkali ions concentration in the pore solution.

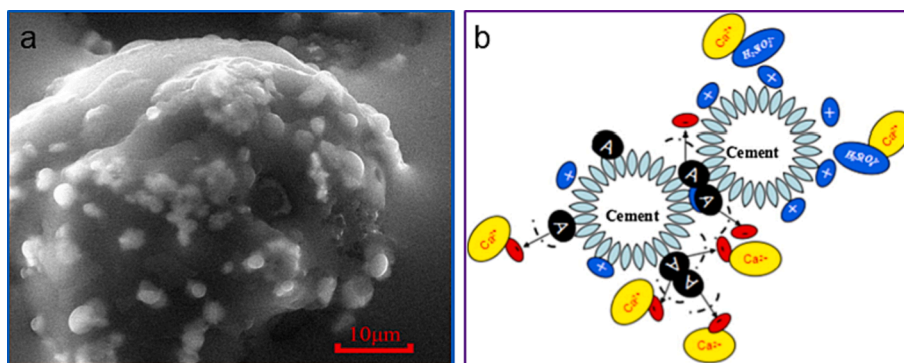


Fig. 6. (a) ESEM images showing the adsorption of anionic AE onto the cement surface (Li et al., 2018); and (b) illustration of the AE membrane on the cement surface (Li et al., 2018).

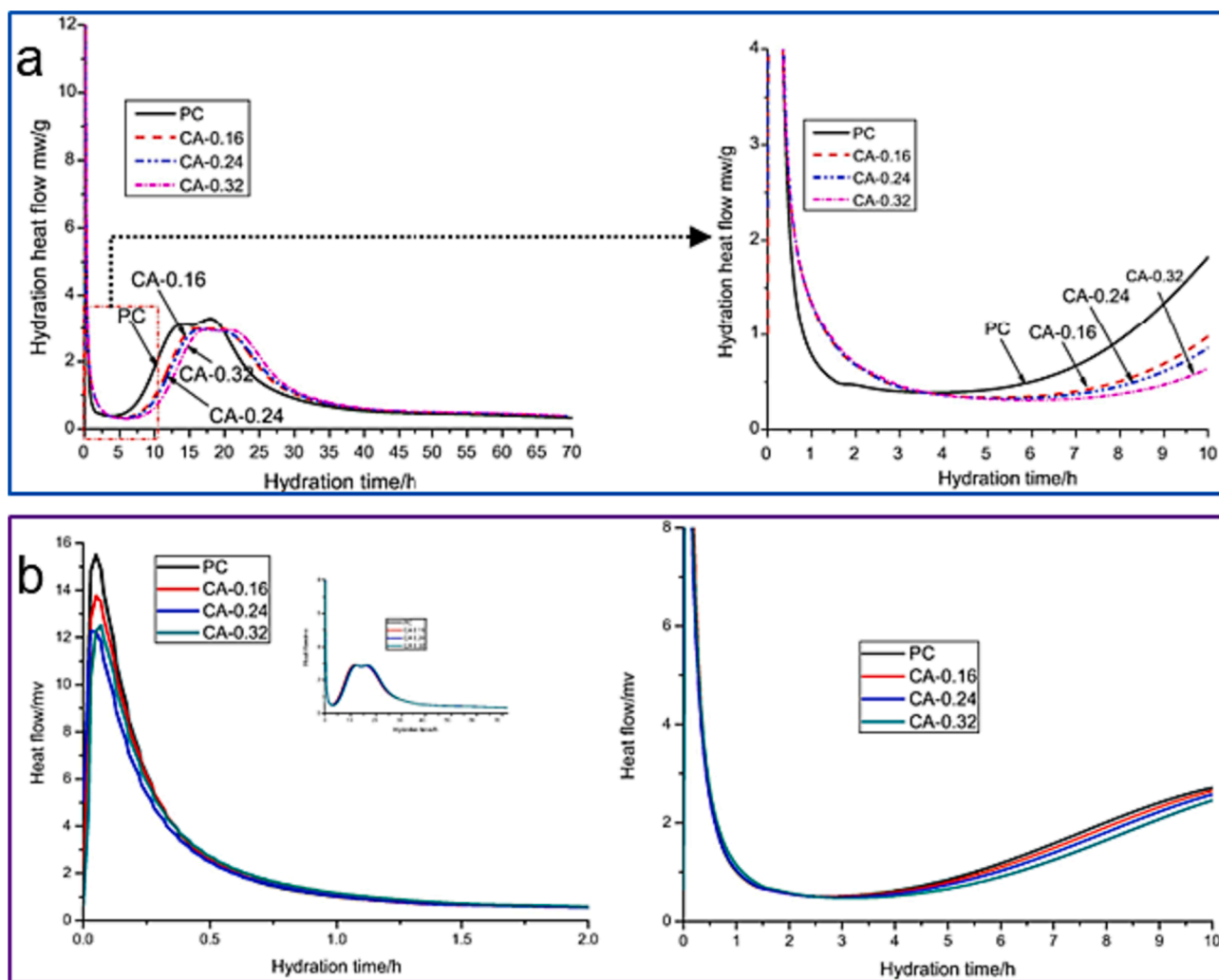


Fig. 7. Influence of (a) anionic AE (Li et al., 2018); and (b) cationic AE on the heat flow of cement hydration (Li et al., 2020).

#### Electrical resistivity changes in the CA system

The release of hydration ions in cement pore solution during cement hydration transforms the cement suspension into a continuous conductive phase (Lu et al., 2022d; Lu et al., 2022f; Ouyang et al., 2016b; Ouyang et al., 2017). The conductivity of the solid-liquid system in the early hydration of cement is related to the concentration of ions, providing a means to measure the dissolution process of cement (Ouyang et al., 2016a; Ouyang et al., 2016c). As shown in Fig. 8, the

conductivity of all CA mixtures initially improves rapidly and then stabilizes with increasing curing time. The distinct phases of conductivity change correspond to the dissolution and induction phases of the hydration process, respectively. The conductivity of the CA paste decreases gradually with increasing A/C (Fig. 8b), indicating that the addition of AE delays the release of hydration ions during the dissolution and induction periods (Ouyang and Tan, 2015; Ouyang et al., 2016d). Comparing the conductivities of CA mortars containing cationic and

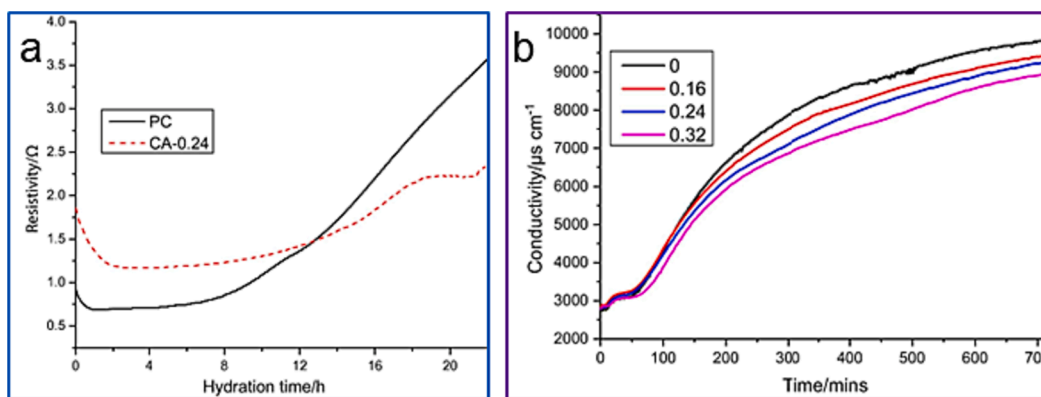


Fig. 8. Influence of (a) anionic AE (Li et al., 2018); and (b) cationic AE on the electrical resistivity of the CA system (Li et al., 2020).

anionic AE for the same hydration time, the cationic AE mortar shows higher conductivity (Ouyang and Tan, 2015; Peng et al., 2015). This can be attributed to the formation of inorganic salts resulting from the interaction of anionic AE with cations. As a result, adding anionic AE into the mixture can result in a decrease in the number of conductive ions, resulting in lower conductivity and less stability.

#### Microscopic investigations in the CA system

Thermogravimetry analysis revealed three distinct endothermic

peaks, including the decomposition of C-S-H gel and Aft, the dehydration of CH, and the decomposition of carbonate, with carbonate primarily formed by the carbonation of CH (Cui et al., 2022; Jiang et al., 2022; Jiang et al., 2020a; Lu et al., 2022e). The intensity of the peaks decreases as increasing A/C (Fig. 9a), indicating a reduction in the total amount of CH produced by cement hydration with the presence of AE. Fig. 9b shows Fourier Transform Infrared Spectroscopy (FT-IR) spectra of CA paste containing different A/C, the intensity or integrated area of the  $960 \text{ cm}^{-1}$  band gradually decreases as increasing A/C from 0 to 0.32,

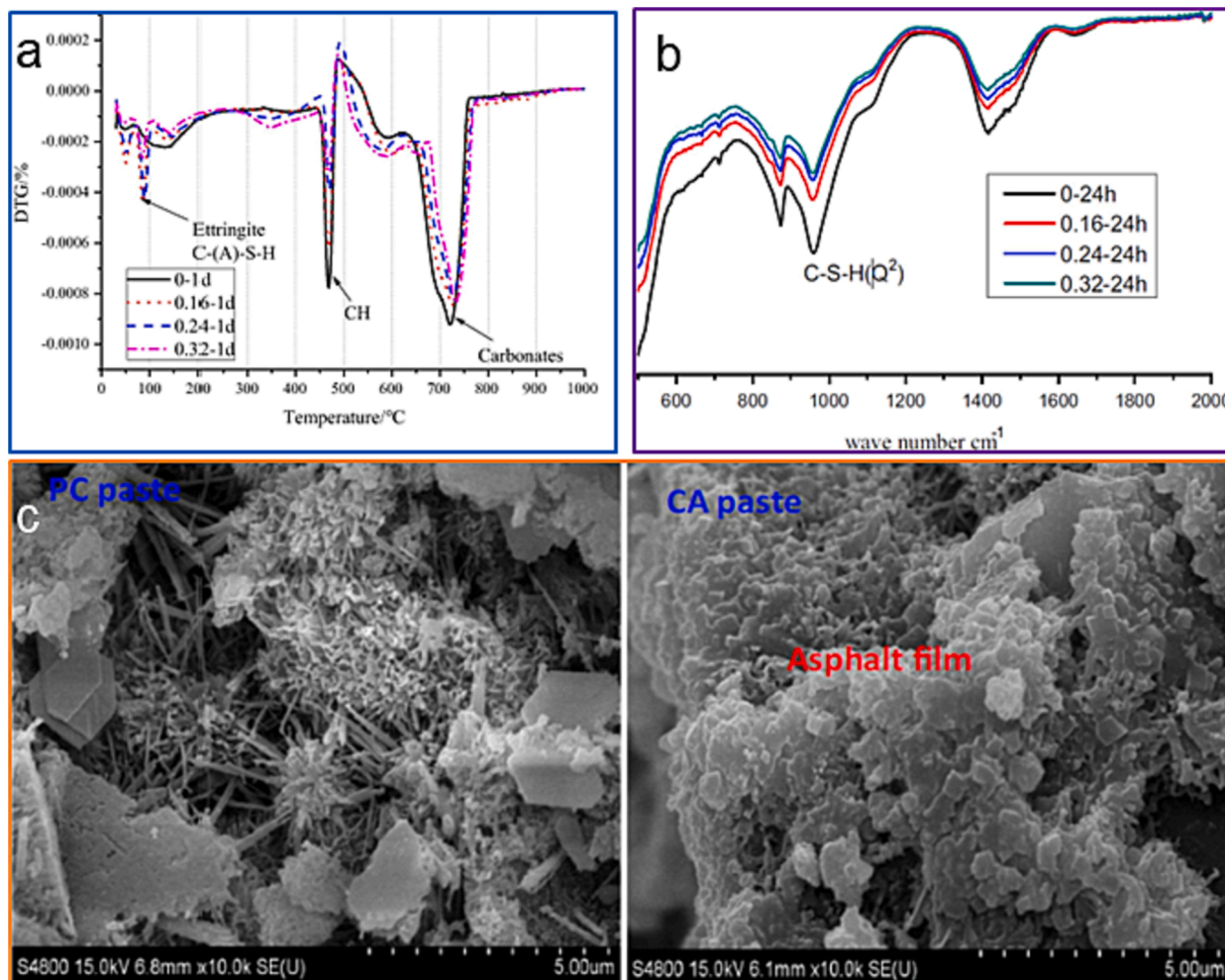


Fig. 9. (a) Thermogravimetry analysis of paste containing different A/C (Li et al., 2020); (b) the variation of the FTIR spectra (Li et al., 2020); and (c) SEM images of CA and cement pastes (Li et al., 2020).



indicating the inhibition of C-S-H formation by the presence of AE. As hydration products form and AE demulsifies, a continuous AE film coats the surface of hydrates, eventually leading to the formation of a cement-AE composite network (Gan et al., 2021; Lu et al., 2021). As suggested in Fig. 9c, the hardened paste comprises amorphous C-S-H, pinpoint Aft, and lamellar CH, with pores full of cement hydrates (Cui and Chang, 2022; Cui et al., 2021; Jiang, T. et al., 2023). The presence of an AE film covering the hydrates' surface, provides evidence of the retardation of cement hydration.

## Performance of CA paste, CA mortar, and CAEM

### Fresh properties of CA paste

#### Setting time

Li et al. (Li et al., 2018) conducted research indicating that the initial and final setting times of CA paste were prolonged as the A/C ratio increased from 0 to 0.32, suggesting that the addition of anionic AE retards cement hydration (Fig. 10a). In a separate study, they found that the initial and final setting times of CA paste with cationic AE increased by 50% and 60% (Fig. 10b), respectively, when the A/C of CA paste increased from 0 to 0.32 (Li et al., 2020). In general, regardless of the type (anionic or cationic) and dosage of AE, the presence of AE led to significantly longer initial and final setting times of CA paste, indicating the inhibition of cement hydration at early stages. It is important to note that in addition to the dosage and type of AE, the water-to-cement ratio (w/c) and curing conditions also influence the setting time of CA paste, as proposed by Tan et al. (Tan et al., 2014).

#### Rheology properties

The adsorption of AE on the cement particles' surface and the changes in the morphology and size distribution of asphalt droplets due to the demulsification of AE can influence the rheological property of CA paste or mortar (Ouyang and Tan, 2015; Ouyang et al., 2016d). Previous studies have shown that the addition of AE has a plasticizing effect on fresh cement paste to some extent (Zhang et al., 2012), as presented in Fig. 11a. The initial yield stress of CA paste reduces as the A/C ratio increases up to 0.2, but it starts to increase when the A/C ratio exceeds 0.2. It is worth noting that even at an A/C of 0.6, the initial yield stress of CA paste is still below cement paste. The rheological properties of fresh CA paste are also influenced by the type of AE (Ouyang et al., 2016b; Ouyang et al., 2018). CA paste containing cationic AE exhibits higher viscosity at low shear rates than anionic AE-modified CA paste, while the apparent viscosity at high shear rates shows the opposite trend (Ouyang et al., 2018; Ouyang et al., 2017; Ouyang and Tan, 2015). Specifically,

when the A/C ratio is 0.40, the viscosity at high shear rates ( $>50 \text{ s}^{-1}$ ) of CA paste with cationic AE is similar to the CA paste incorporating anionic AE. However, as the A/C ratio increases to 1.40, the viscosity at high shear rates ( $>20 \text{ s}^{-1}$ ) of CA paste containing cationic AE becomes much lower than that of CA paste incorporating anionic AE (Ouyang and Tan, 2015). These findings suggest that the difference in viscosity of the mixture at high shear rates of CA pastes with different types and dosages of AE (Ouyang et al., 2017), as shown in Fig. 11b.

The addition of AE to CA paste results in the adsorption of AE on the cement surface, leading to a decrease in strength and shear-thinning viscosity (Ouyang et al., 2016b; Ouyang et al., 2017; Ouyang et al., 2016d). The presence of anionic AE reduces the shear thinning response and viscosity of CA paste compared to cationic AE. The incorporation of a superplasticizer helps prevent the flocculation of cement and asphalt droplets, thereby prolonging the demulsification process of AE in CA paste or CA mortar (Ouyang et al., 2016b). Furthermore, factors such as the concentration of  $\text{Ca}^{2+}$ , pH value, particle volume fraction, and resting time also affect the rheological performance of CA paste or CA mortar (Ouyang et al., 2018; Ouyang and Tan, 2015; Ouyang et al., 2016d; Peng et al., 2015; Tan et al., 2014).

### Mechanical properties of CA mortar and its application in ballastless slab track

The hardened paste and AE in the CA system exhibit distinct mechanical and viscoelastic properties (Buczyński and Iwański, 2017; Castañeda López et al., 2018; Chen et al., 2020). The overall mechanical strength of hardened paste or mortar varies significantly with changes in the A/C ratio (Ayar, 2018; Bołtryk and Małaszkiwicz, 2013; Buczyński, 2016). In the context of ballastless slab tracks, which are widely utilized in high-speed railways all over the world, CA mortar plays a crucial role as a damping material between the rigid slab and concrete base, offering advantages such as long service life and low maintenance (Fang, X. et al., 2016; Gao et al., 2012). Therefore, this section will review and discuss the static and dynamic mechanical properties of CA mortar in the context of its application in ballastless slab tracks (Rutherford et al., 2014; Sun et al., 2019; Wang et al., 2015).

#### Static mechanical behaviour

The compressive properties of CA mortar primarily rely on the cement hydrates (Diaz, 2016; Dolżycki et al., 2017; Dolżycki et al., 2017). Typically, as the A/C increases, it leads to higher air voids in the CA mortar. Moreover, the formation of hydration products from cement decreases, leading to a decrease in the mechanical strength of CA mortar (Liu et al., 2014; Moghadas Nejad et al., 2017). In addition, increasing

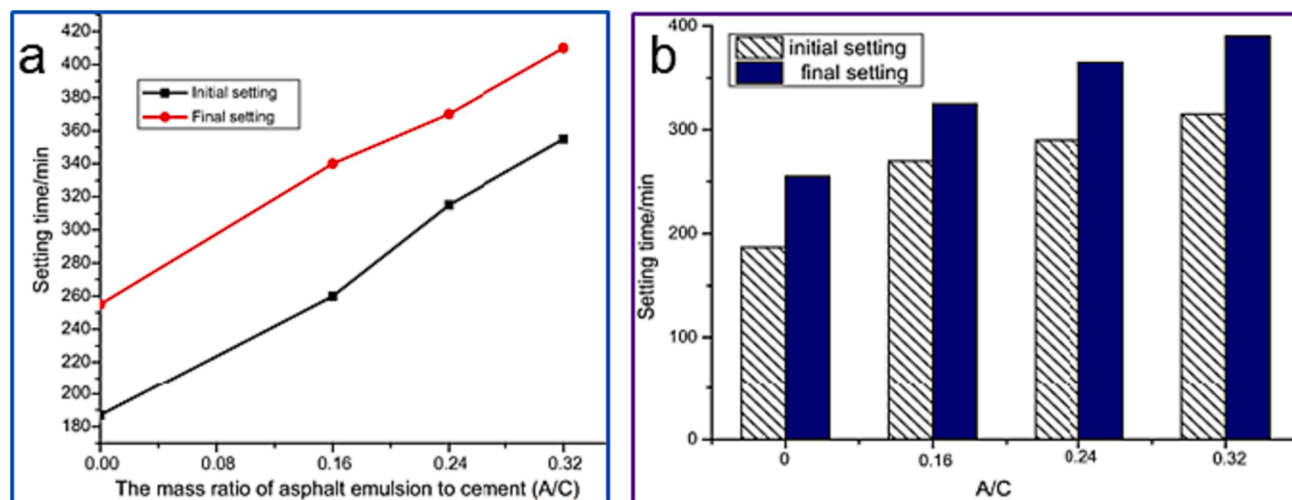


Fig. 10. Influence of (a) anionic AE (Li et al., 2018); and (b) cationic AE on the setting times of CA pastes (Li et al., 2020).

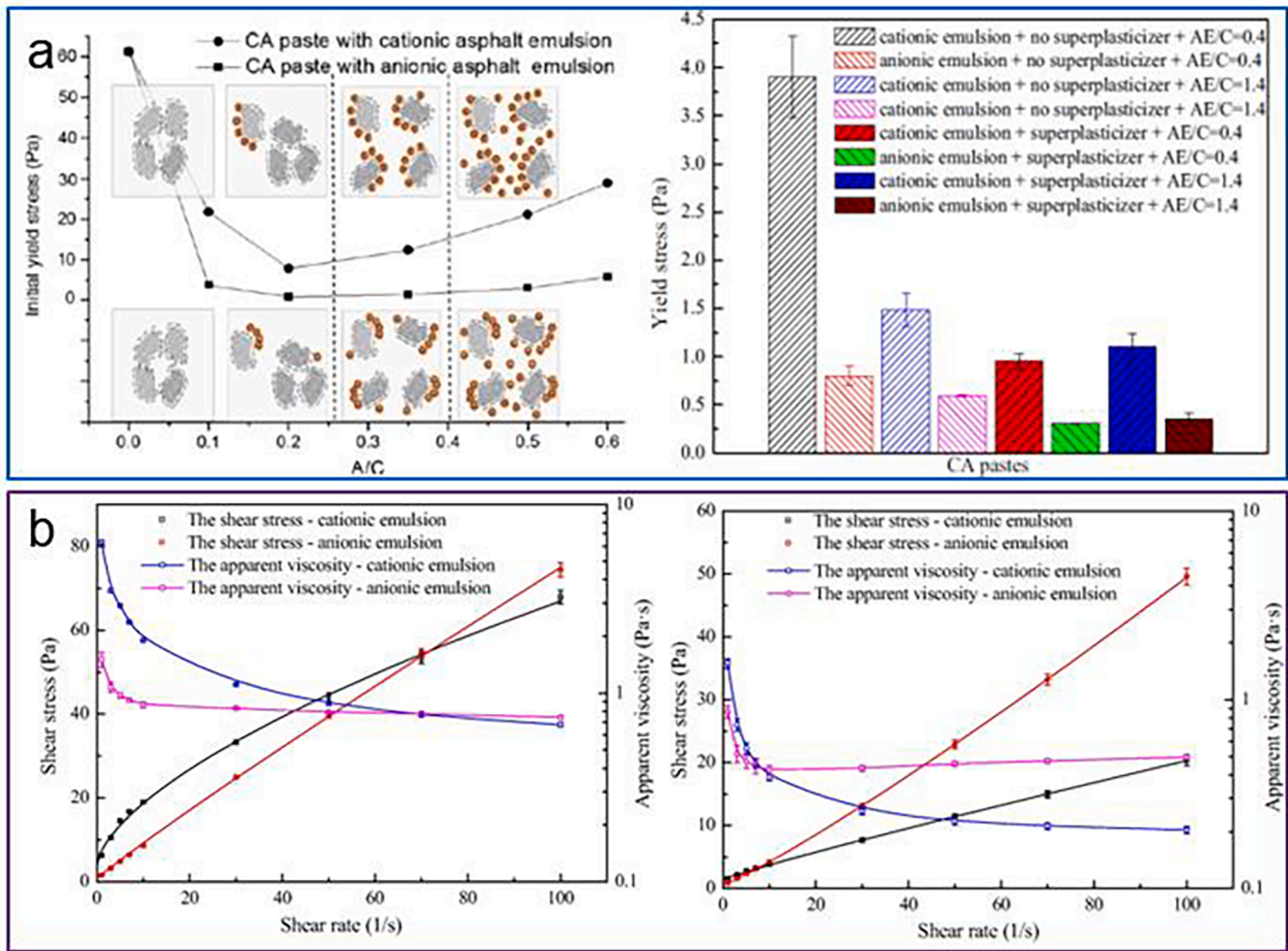


Fig. 11. (a) Initial yield stress of fresh CA paste (Zhang et al., 2012); and (b) the influence of AE types on the rheological performance of CA paste (Ouyang et al., 2017).

the A/C can create weaker regions at the interfaces between organic (AE)-inorganic (cement hydrates) phases and asphalt-fine aggregates, which can contribute to the decrease in compressive strength of CA mortar (Dong et al., 2018; Doyle et al., 2013). Experimental studies conducted by Liu et al. (Liu et al., 2014) on statically loaded CA mortar demonstrated that the modulus of elasticity and damage energy all decrease with increasing A/C [62]. Similarly, Liu et al. (Liu and Liang, 2017) suggested that the compressive strength of CA mortar decreases with increasing A/C but increases with curing age. This is because of the reduction in cement hydrates, the hindrance of cement hydration by AE, and the increase in air content within the CA system. Furthermore, the demulsification of AE results in the formation of an AE film that coats the surface of unhydrated cement, impeding their further hydration. This phenomenon, known as the retarding effect of AE, becomes more pronounced with higher dosages of AE in CA mortar.

There are two typical kinds of CA mortars: Type I (low-modulus CA mortar) used for slab track and Type II (high-modulus CA mortar) used for continuous slab applications (Deng et al., 2021; Gao et al., 2012; Wang et al., 2015). Interestingly, these two types of CA mortars exhibit distinct mechanical characteristics due to their differing A/C ratios (Qin et al., 2022; Rutherford et al., 2014). In the case of a relatively low A/C, CA mortar behaves as a brittle composite, where the hardened paste acts as the continuous phase forming a skeletal structure. This results in overall mechanical strengths similar to those of pure hardened cement mortar, with the asphalt serving as a secondary binder filling the matrix.

However, as the A/C increases, the material undergoes a transition from quasi-brittle fracture to ductile fracture. This increase in fracture toughness enhances the damping ratio of the slab track system [65]. When the A/C ratio is relatively large, CA mortar exhibits the characteristics of a ductile material. This network structure, however, is relatively weak, leading to a significant reduction in the mechanical strengths of CA mortar. As suggested by Wang et al. (Wang et al., 2015), as A/C is below 0.50, the CA mortar displays quasi-brittle fracture behavior. Conversely, the CA mortar exhibits ductile fracture behavior as the A/C increased to 0.50.

#### Dynamic mechanical behaviour

CA mortar serves as a resilient pad in high-speed railroad slab tracks (as shown in Fig. 12a). Consequently, it is inevitably subjected to different load rates, and its response to dynamic loading is crucial for its proper functioning within the slab track. Various factors, including the A/C ratio, loading frequency, loading rate, and temperature, significantly influence the dynamic mechanical properties of CA mortar. These factors collectively determine the performance and effectiveness of CA mortar in the slab track.

For instance, Fu et al. (Fu et al., 2017) conducted a study on the energy release of CA mortar and investigated the relationship between the energy release coefficient and AE content. The energy release coefficient serves as an indirect measure of the damping ratio of CA mortar. The researchers observed that as the AE content increased, the energy

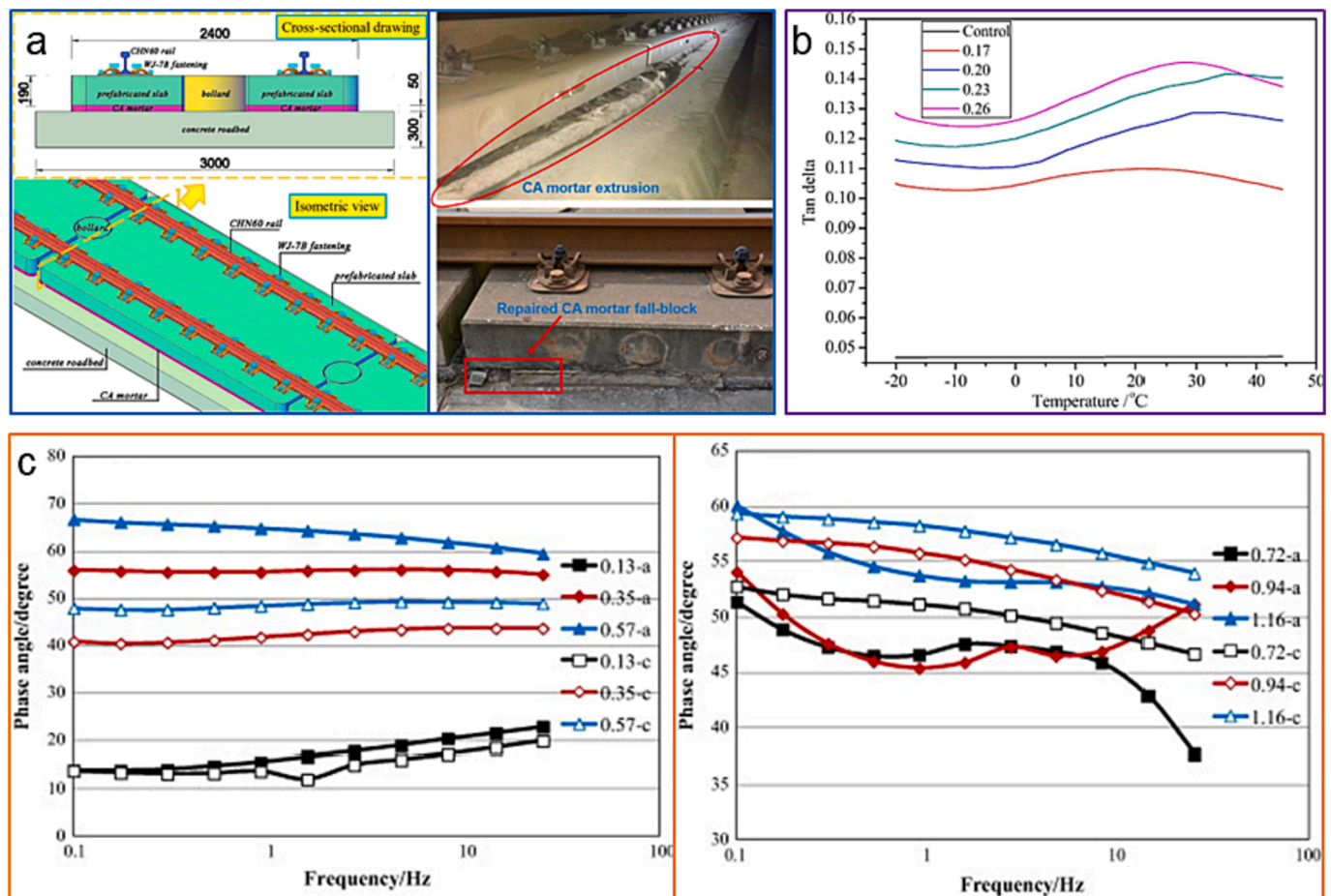


Fig. 12. (a) The CRTS I ballastless track and CA mortar disease (Deng et al., 2021); (b) loss factor changes of the mortars (Leiben et al., 2018); and (c) phase angle of CA pates (Wang et al., 2015).

release coefficient gradually rose, indicating accelerated material energy dissipation and an increase in the damping ratio (Deng et al., 2021; Wang et al., 2015). Leiben et al. (Leiben et al., 2018) also suggested that adding AE can enhance the energy dissipation capacity of CA mortar. Specifically, for CA mortar with increasing A/C to 0.17, the viscoelasticity of the mortar was found to be poor, with the loss factor increasing with increasing A/C (Fig. 12b). Wang et al. (Wang et al., 2015) suggested that in high-strength CA mortar, the shear modulus decreases with increasing A/C ratio at low loading frequencies, whereas the opposite trend is observed in low-strength CA mortar (Fig. 12c). This discrepancy can be attributed to the difference in AE content. Moreover, the phase angle increases with the rising A/C ratio in both low and high-strength CA mortar, indicating a greater significance of the viscoelastic properties of the mortar [50,62]. Wang et al. (Wang et al., 2015) further examined the influence of loading rate on the properties of CA mortar at low A/C ratios. The results revealed that the peak stress increases with increasing loading frequency within the range of 1 mm/min to 20 mm/min, while it remains nearly constant between 20 mm/min and 50 mm/min.

Based on the aforementioned findings, it is observed that the compressive strength of CA mortar decreases as increasing the A/C. However, this reduction in strength is accompanied by increased ductility and decreased brittleness (Tian et al., 2020). The fracture mode of CA mortar shifts from quasi-brittle to ductile, exhibiting viscoelastic behavior. Therefore, it is crucial to carefully regulate the A/C of CA mortar based on specific application requirements. Furthermore, the addition of AE proves to be an effective method of enhancing the damping characteristics of CA mortar, making it suitable as a damping

material in ballastless slab tracks.

#### The road performance of CAEM and its applications in pavement construction

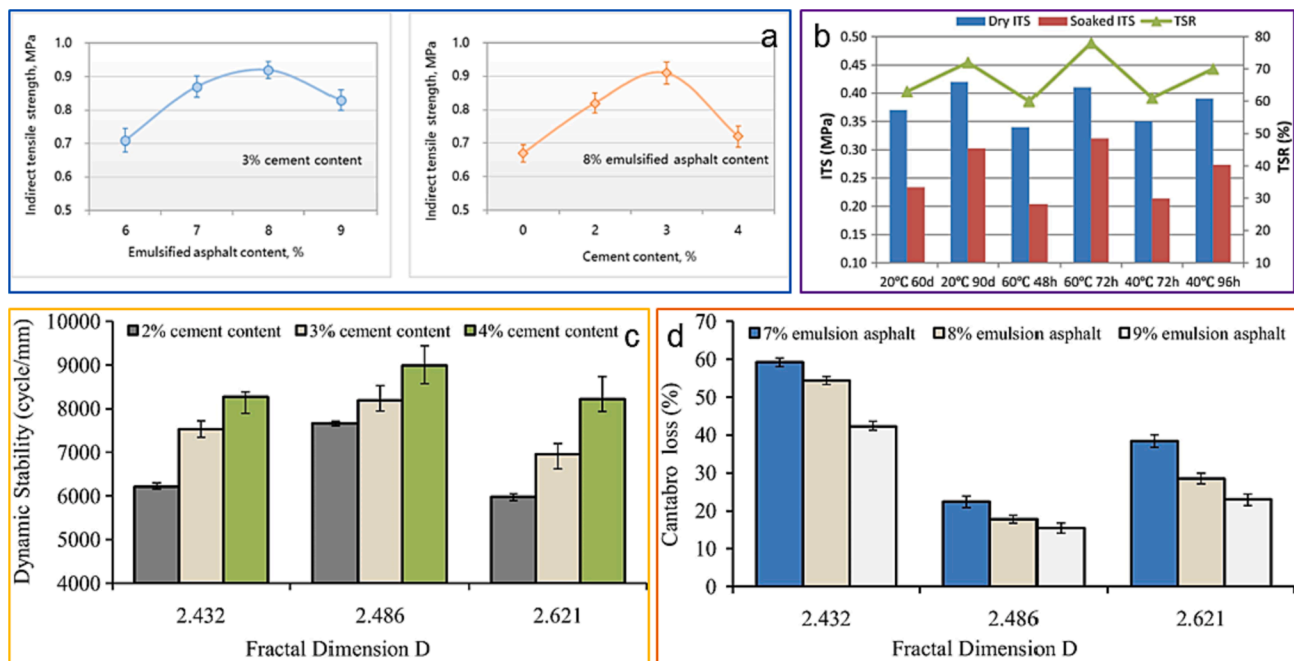
As a type of inorganic–organic composite, Cement Asphalt Emulsion Mixture (CAEM) is typically composed of cement, AE, aggregate, and chemical additives, it has higher compressive strength compared with asphalt mixture and superior flexibility than cement concrete, enabling it has been extensively applied in pavement construction and maintenance (Boitryk and Małaszkiwicz, 2013; Tian et al., 2020; Wang et al., 2015). As such, this section focuses on reviewing and discussing the road performance of CAEM.

#### Mechanical properties

The mechanical properties of CAEM can be assessed through various parameters such as indirect tensile strength, compressive strength, Marshall Stability, and modulus of resilience (Xiao et al., 2019; Zhu et al., 2019a; Zhu et al., 2019b). Adding cement to CAEM can enhance its mechanical strength and it is primarily attributed to the cement hydrates in the composite, which increases the stiffness of the mixture (Yan et al., 2017; Yan et al., 2014; Yang et al., 2021).

In research performed by Fu et al. (Xiao et al., 2019) and Tian et al. (Tian et al., 2020), it was observed that the indirect tensile strength and modulus of resilience of CAEM initially improved and then reduced when the AE content increased from 6 wt% to 9 wt% while keeping the cement at 3 wt%, as depicted in Fig. 13a. This indicates that there exists an optimal amount of cement for enhancing the mechanical strengths of





**Fig. 13.** (a) Affect of cement and AE content on the indirect tensile strength of CAEM (Xiao et al., 2019); (b) soaked TSR results of CAEM (Du, 2018); (c) relationship of dynamic stability and fractal dimension (Xu et al., 2017); and (d) relationship of Cantabro loss index and fractal dimension (Xu et al., 2017).

CAEM. The choice of cement type also influences the mechanical strengths of CAEM, primarily due to variations in hydration rates and microstructure optimization associated with different cement types (Thanaya et al., 2014; Wang et al., 2018; Xie et al., 2019). For instance, Saadoun et al. (Saadoun et al., 2018) demonstrated that CAEM modified with calcium sulphoaluminate cement (CSA) exhibited the highest stability compared to calcium aluminate cement (CAC). This can be attributed to CSA producing larger quantities of the binder during hydration. The difference in binder production contributes to the improved mechanical strengths of CSA-modified CAEM. Moreover, aggregate gradation is another factor that affects the mechanical strengths of CAEM. The mechanical strengths of CAEM are related to the fineness modulus of aggregates. It has been found that an optimum gradation, characterized by an appropriate fractal dimension value ( $D$ ), helps achieve a balance between the framework and density of CAEM for a given binder dosage (Xu et al., 2017).

Indeed, the addition of cement can enhance the mechanical strengths of CAEM, but it is important to determine the optimal amount of cement for the specific CAEM system. Along with the A/C ratio, other factors such as curing temperature and moisture content also play crucial roles in determining the early-stage strength of CAEM (Lin et al., 2018). Further research is needed to investigate and unlock the key factors that influence the early-stage strength of CAEM, providing valuable insights for optimizing the curing conditions and achieving desired mechanical properties. Exploring these factors in future studies will contribute to the continued improvement and development of CAEM.

#### Moisture susceptibility

Moisture can affect the performance of CAEM, as it can weaken the bonding strength between AE and aggregates and lead to asphalt stripping from the aggregate surface (Al-Khateeb and Al-Akhras, 2011; Ayar, 2018; Boltryk and Malaszkiewicz, 2013; Dulaimi et al., 2016). The moisture present in aggregates, pre-mix water, and AE all contribute to the overall moisture resistance of CAEM (Buczyński, 2016; Buczyński and Iwański, 2017; Castañeda López et al., 2018; Du, 2018). Notably, CAEM-containing cement has shown improved moisture resistance compared to cold mixtures without cement, as evidenced by high soaked indirect tensile strength ratio (TSR) and freeze-thaw (FT) TSR values,

exceeding 90% and 75%, respectively (Fig. 13b) (Du, 2018). The presence of cement in CAEM enables a reaction with external moisture and water from AE demulsification, resulting in the formation of cement hydrates. These hydrates interlace with the AE film, creating a grid structure that enhances the moisture resistance of CAEM (Xiao et al., 2019). Interestingly, a study reported that when the cement content in CAEM reached 2.5 wt%, the soaked TSR and FT-TSR even exceeded 100%, mainly due to the activation of cement hydration under long-term water conditions (Du, 2016). Another study by Tian et al. (Tian et al., 2020) confirmed that adding higher cement contents increases the moisture resistance ability of CAEM, as the increased cement content forms more hydrates and enhances the stiffness of the mixture. Recent research has also explored the positive effects of microwave radiation on enhancing the water stability of CAEM. Microwave heating accelerates cement hydration, resulting in the formation of a denser microstructure, thereby enhancing the moisture resistance of CAEM (Wang et al., 2020). Furthermore, the moisture susceptibility of CAEM may be influenced by factors such as aggregate gradation and curing regime, which warrant further investigation in future research.

#### High-temperature performance

Rutting has emerged as a significant distress in asphalt pavements, as they are prone to permanent deformation under the influence of load and high temperatures. If timely maintenance is not carried out, the accumulated deformation can lead to structural damage to the pavement (Castillo et al., 2015; Liu et al., 2021; Liu et al., 2022a; Rafiq Kakar et al., 2022; Zhou et al., 2022). However, adding cement to CAEM can mitigate the temperature sensitivity and enhance its high-temperature viscosity, thereby improving rutting resistance (Tian et al., 2020). Research by Xiao et al. (Xiao et al., 2019) demonstrated that the high-temperature rut resistance of CAEM decreased with increasing A/C when the cement content was fixed at 3%. Similarly, Xu et al. observed a positive effect of cement addition on enhancing the rutting resistance of CAEM and identified the optimal cement content to be between 2% and 3% (Xu et al., 2017). Conversely, the dynamic stability of CAEM containing 9 wt% AE was significantly lower than that of CAEM with 8 wt% AE. This can be attributed to the higher amount of AE leading to a greater likelihood of aggregate slippage and flow, ultimately reducing the rutting

resistance at high temperatures (Xiao et al., 2019). In Fig. 13c, Xu et al. (Xu et al., 2017) demonstrated that the dynamic stability (DS) of CAEM is influenced by the fractal dimension. Among the three gradations considered, G2 with a reasonable fractal dimension exhibited the highest DS values regardless of the cement content, because the aggregate gradation and the adhesion of CAEM jointly determined its high-temperature stability (Xu et al., 2017).

#### Low-temperature performance

In cold regions, low-temperature cracking is a significant concern for asphalt pavements (Shi et al., 2022; Xiao et al., 2019; Xu et al., 2021; Yu et al., 2023). Research by Yan et al. (Yan et al., 2017) revealed that Mix I, which did not contain cement, exhibited the lowest failure strain. However, the failure strain initially increased and then decreased as increasing the cement content. Among the four mixtures, Mix III, containing 1.5% cement, demonstrated the best resistance to low-temperature cracking. This suggests that adding cement to CAEM can enhance its resistance to low-temperature cracking, but there may be an optimal cement content that yields the best results (Yan et al., 2017). Similarly, Tian et al. (Tian et al., 2020) observed that adding cement to CAEM improves its toughness and strength, which in turn enhances the cohesive force of the asphalt binder. This improvement in shear strength contributes to the mixture's performance at low temperatures and enhances its resistance to cracking. Additionally, a higher AE content generally leads to a greater increase in cracking resistance at low temperatures (Wang et al., 2014). The cracking resistance of CAEM also improves with increased curing age and temperature. For example, the cracking resistance gradually increases with longer microwave radiation time, indicating that microwave radiation effectively enhances the crack resistance of CAEM at low temperatures (Wang et al., 2020).

#### Cantabro loss

The Cantabro abrasion test was employed to assess the anti-abrasion property of the mixture under impact load (Xiao et al., 2019). In general, higher cement content in CAEM results in increased cohesion force and reduced ravelling loss rate, suggesting that introducing cement positively impacts the anti-abrasion ability of CAEM (Yan et al., 2017). Similarly to the improvement in low-temperature performance, the Cantabro loss value initially decreases and then slightly increases as increasing the cement dosage from 0% to 4% (Xiao et al., 2019). Furthermore, increasing AE content can enhance the abrasion resistance of CAEM with 3 wt% cement. As the AE content increases to 8%, the asphalt film enveloping the aggregates thickens, thereby enhancing the integrity and cohesiveness of CAEM and maintaining the Cantabro loss of the mixture below 20%. Xu et al. (Xu et al., 2017) observed significant differences in the Cantabro loss value among different aggregate gradations while keeping the cement content constant (Fig. 13d). Additionally, the Cantabro loss decreases as the AE dosage increases, regardless of the gradation type. Overall, the Cantabro loss of CAEM is closely related to its strength. Enhancing the strength of CAEM ensures its integrity and cohesion, thereby reducing Cantabro loss. It is important to establish the relationship between strength, particularly early-age strength, and Cantabro loss of CAEM. Future studies can focus on using the early-age strength of CAEM to predict Cantabro loss and further understand the relationship between strength and anti-abrasion performance.

#### Conclusions and prospectives

This paper presents a comprehensive review of the interactions between organic-inorganic composites and their influence on the performance of Cement Asphalt Emulsion Mixture (CAEM). Firstly, we delve into the interactions between asphalt emulsion (AE) and cement, highlighting their significance in the CAEM system. Subsequently, we analyze the demulsification of AE and cement hydration, elucidating their effects on the performance of CAEM. Finally, we discuss the

performance of CAEM, considering various factors and drawing important conclusions and prospects from our analysis. The main findings of this review can be made as follows.

(1) The interaction between AE and cement encompasses a complex interplay of physical and chemical processes, including the adsorption of AE onto the cement surface and the destabilization of AE. These interactions are influenced by three key factors: the type and dosage of the AE, as well as the A/C ratio. Note that different mineral components present in various types of cement exhibit varying capacities for adsorbing AE. Therefore, conducting an in-depth investigation into the competitive adsorption behavior among mineral components can provide valuable insights into achieving controllable adjustments of compatibility in various combinations of cement and AE.

(2) The retardation mechanisms of AE during cement hydration can be ascribed to several factors: a) the adsorption of AE on the cement surface, b) the formation of a coating asphalt film through demulsification of AE, and c) the formation of calcium complexes. It has been observed that the retarding effect becomes more pronounced with higher A/C ratios, regardless of the type of AE employed. However, the current focus on the retardation effect of AE during cement hydration primarily revolves around the adsorption quantity of AE droplets onto the cement surface, while the coverage area of AE on cement surfaces has been overlooked. In future research, it is essential to delve deeper into the understanding of the roles played by AE during cement hydration. Such investigations will provide valuable academic support for the development of next-generation CAEM.

(3) Adding anionic AE can result in lower yield stress and viscosity of CA paste relative to the cationic AE. The anionic AE can be used to enhance the fluidity of CAEM with low A/C, whereas CAEM containing high A/C should be fabricated by applying cationic AE to ensure early-age strength. Adding superplasticizers can prevent the flocculation of cement and asphalt droplets, thus prolonging the demulsification process of AE in CAEM. Additionally, the concentration of  $\text{Ca}^{2+}$ , pH value, particle volume fraction, and resting time all impact the rheological performance of CA paste or CA mortar.

(4) The compressive strength of CA mortar reduces with increasing the A/C, while it acquires higher ductility and lower brittleness. Typically, its fracture mode transitions from quasi-brittle fracture to ductile fracture. As such, the A/C ratio of the CA mortar should be strictly regulated according to the requirement of practical applications. Additionally, adding AE can effectively improve the damping characteristics of CA mortar and can be used as a damping composite in ballastless slab tracks.

(5) Introducing cement in CAEM has a positive impact on various aspects of its performance. It improves mechanical strengths, moisture susceptibility, high-temperature performance, low-temperature performance, and Cantabro loss. However, it is important to note that there exists an optimal amount of cement to be used in the CAEM system. The A/C ratio, along with the suitable curing temperature and moisture content, play significant roles in determining the early-stage strength of CAEM. Further research should focus on identifying the key factors influencing early-age strength to optimize the performance of CAEM.

#### Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

#### Data availability

Data will be made available on request.

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