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Review Article

Microstructure in Adhesion Development Process and Strength Formation Mechanism in Early Stage of Cold Recycled Mixture with Emulsified Asphalt

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With the large-scale maintenance and renovation of asphalt pavement, a considerable amount of reclaimed asphalt pavement (RAP) will be generated. Stacking these wastes occupies a large amount of land and seriously damages the ecological environment. Hence, efficient regeneration of RAP through recycling technology has gained more and more attention. In this paper, some topics were reviewed to further promote the cold recycling (CR) technology and better follow-up the research progress. First, it discussed the raw materials and the differences and similarities between cement-emulsified asphalt mortar (CEAM) and cold-recycled mixtures with emulsified asphalt (CRME). Second, it reviewed the adhesion development of emulsified asphalt mastic, the application of X-ray technology in microscopic study of CRME and the characteristic of strength development of CRME. The adhesion development of CRME begins with the process of demulsification and hydration caused by the migration and dissipation of water inside the emulsified asphalt mastic. In addition, many factors would influence this process simultaneously. However, the microbehavior mechanism of internal water transport in emulsified asphalt mortar has not yet been thoroughly revealed, and it lacked scientific measurement research on the promoting effect of complex conditions on the development of adhesive properties of mixtures. Therefore, in this paper, they were suggested for future research.

1. Introduction

Since the rapid development of road industry worldwide, especially in the highway engineering, the asphalt pavement has become an important form of pavement structures. In some countries, the application proportion of asphalt pavements have exceeded 95% of all highway pavements [1]. However, there is a significant reduction in the service life of asphalt pavements of some roads, and it usually cannot reach the design life, due to the limited level of transportation management, the frequent overloading and heavy loading, the lack of high-quality road materials and deficiencies in the construction quality control system. At the same time, an asphalt pavement is prone to aging under natural conditions, such as ultraviolet light, hydroxyl radical, oxygen, ozone, nitrogen oxide and temperature, which exacerbates the deterioration of its performance. Hence, after 5–6 years of operation, it enters a period of large-scale maintenance and renovation.

At present, the maintenance and renovation process of asphalt pavements mainly involves milling the old asphalt layer and overlaying a new asphalt layer, which would generate a considerable amount of reclaimed asphalt pavement (RAP). According to statistics, the amount of RAP produced is ∼180 million tons every year only in China [1]. Stacking these wastes would occupy a large amount of land and seriously damage the ecological environment. In addition, overlaying a new asphalt layer results in a severe shortage of high-quality asphalt and mineral materials, and causes a significant burden on the allocation and utilization of resource. Therefore, achieving systematic and efficient regeneration of RAP would not only save resources and alleviate the shortage of highway maintenance funds but also conform to the concept of low carbon and environmental protection. It is an important technical

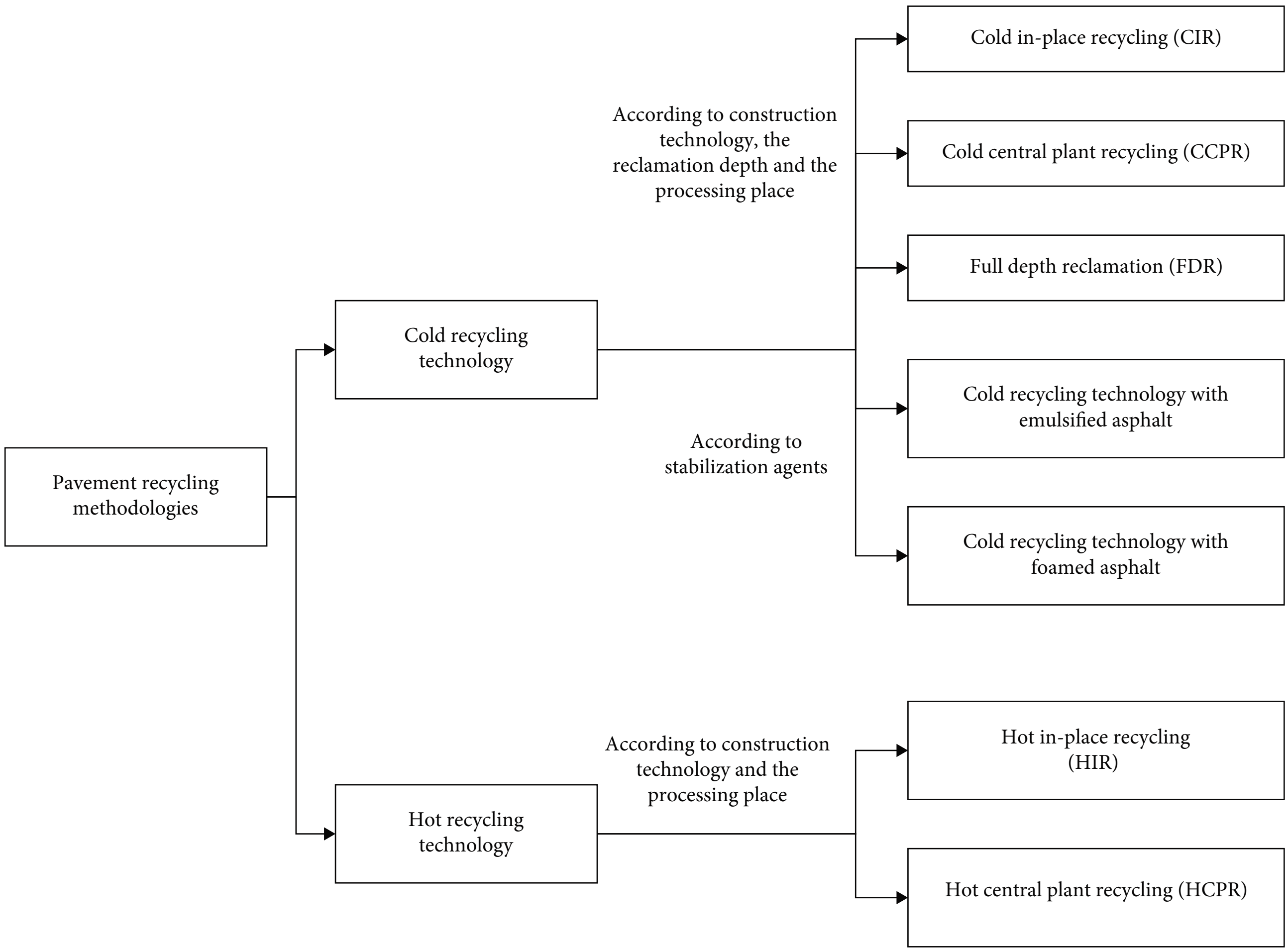


Figure 1: Classification of pavement recycling methodologies.

approach to promote the sustainable development of highway construction [2].

According to the mixing temperature, recycling methodologies can be classified as hot recycling (HR) and cold recycling (CR) as shown in Figure 1. In HR technology, the heating temperature ranges from 120to 160°C, and the RAP content is relatively low, which is generally 10%–40% [3–7]. However, in CR technology, the construction process is completed under natural conditions without heating. Hence, it would greatly avoid the consumption of fossil fuels during the hot regeneration process, resulting in the decrease of pollutant emission such as CO_2, CO, and NO, etc. and the reduction of environmental pollution and builder health damage [8–12]. In addition, all of the RAP materials could be reused in new blended mixture theoretically [13–15]. Therefore, CR technology becomes more and more popular.

According to the usage of the asphalt stabilization agents, CR technology can be classified as CR with emulsified asphalt (CRME) and CR with foamed asphalt (CRMF). Usually, 1.5%–2.5% cement would be added in CRME as an activator to accelerate the emulsion breaking and improve the comprehensive performance of asphalt-stabilized marginal materials, while cement would not be added in CRMF. In engineering practice, both CRME and CRMF have been applied widely. Moreover, CRME has gained more attention because its mixing temperature is 20–70°C, while the mixing temperature of CRMF before foaming is 160–180°C [3, 16, 17], resulting in the less energy consumption of CRME. Hence, in this paper, it will mainly focus on CRME.

Cold recycled mixtures with emulsified asphalt (CRME) is a multiphase composite and a thermodynamically incompatible semi-loose system mainly composed of emulsified asphalt evaporation residue, cement hydration products, RAP materials, new aggregates, and water [18]. In engineering practice, there is another material called cement-emulsified asphalt mortar (CEAM). CEAM is mainly applied in the track structure of high-speed railways, and it is an intermediate layer flung within the space between the track slab and the trackbed [19–22]. In spite of the difference in mixture component ratios and structural capacity, CRME and CEAM are very similar. They both use emulsified asphalt and cement as the binder and consist of emulsified asphalt, aggregates, cement, and water. Hence, when conducting research and evaluation on CRME, CEAM could also be considered, especially research on the mechanism of material strength formation.

Studying the mechanism of strength formation in CRME started from the material preparation. To achieve the paving and compaction, water was added to produce emulsified

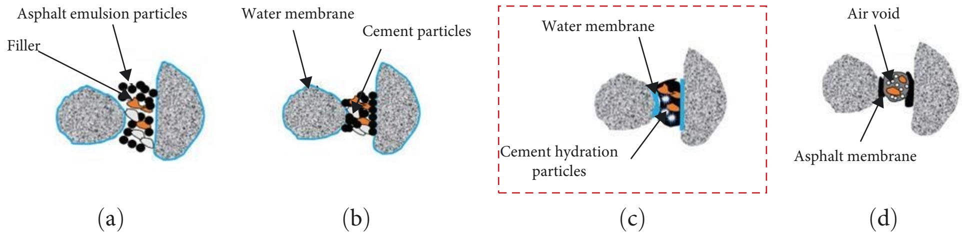


Figure 2: Schematic diagram of microstructure formation of CRME; (a) Mixing process, (b) after compaction, (c) early-stage curing, (d) long-term curing [26].

asphalt and to reduce the viscosity under the natural condition. Hence, it took a period for water to migrate and dissipate. In this process, the adhesion of emulsified asphalt would increase. Meanwhile, the released water would be used for cement hydration and the generated hydration products could improve the early strength of CRME [23]. In this way, the strength of CRME developed gradually. Relevant studies have shown that the early strength development of CRME was significantly positively correlated with the long-term performance [24, 25]. Therefore, to improve the long-term performance of cold recycled mixtures, it was essential to conduct research on the early strength development mechanism of CRME. One of the most important types of research is the microstructure in adhesion development process and strength formation mechanism.

In summary, to further promote the application of CRME in practice, and better follow-up the research progress, this paper reviewed: (1) raw materials of CRME; (2) the similarity and difference between CEAM and CRME; (3) the adhesion development of emulsified asphalt binder in the water migrating and dissipating process; (4) the application of X-ray technology in microscopic study of CRME; and (5) the strength development of CRME.

2. Raw Materials

CRME mainly consists of RAP, emulsified asphalt, cement, and water. The early strength development is completed by the combined action of raw materials, and this progress is shown in Figure 2.

2.1. RAP Materials. RAP materials are mainly composed of aggregates wrapped with aged asphalt. Traditionally, the aged asphalt was ignored, and RAP materials could be considered as "black rock," which acted as skeleton in the mixture. However, although the CRME did not undergo the hot regeneration process, the new and aged asphalt would still be blended to a certain extent [27–29]. It was possible that the aged asphalt should not be ignored, and it is an important factor for evaluating the performance of RAP materials [27]. Therefore, it is of significant engineering value to test the content and related indicators of aged asphalt on the surface of RAP materials.

2.2. Emulsified Asphalt. Emulsified asphalt is formed by insoluble hot melt asphalt sheared and grinded at high speed through a colloid mill, and the asphalt would be dispersed in a fine particle state in an aqueous solution containing the emulsifier, forming an oil in water emulsion. Asphalt is the dispersion phase, and water is the dispersion medium. After the mixing of emulsified asphalt, RAP and other raw materials, CRME is paved and compacted. Under the physical and chemical effects of water evaporation aggregate adsorption, etc., the properties of emulsified asphalt would change. Asphalt would be separated from the water phase in the emulsion, and small asphalt particles would be coalesced to form a continuous integral membrane, which is called the demulsification of emulsified asphalt. After the demulsification, the newly formed asphalt would be wrapped around RAP materials, and the aged asphalt and new asphalt would be merged to form a new asphalt membrane [30].

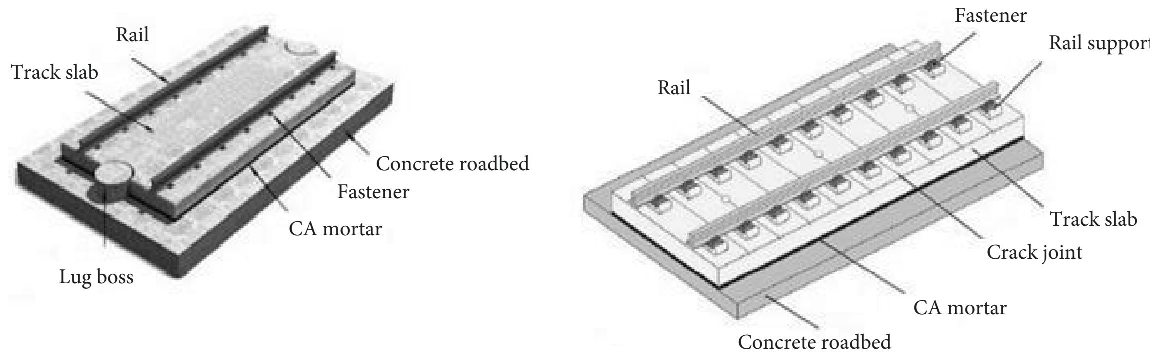
2.3. Cement. To improve the mechanical properties of CRME, an appropriate amount of cement would be considered in the CR project. In the mixture, a part of cement would undergo hydration reaction after absorbing water, which would accelerate the rate of strength development in the early stage. Other cement that does not react with water would be used as filler to adjust the gradation of CRME [15]. Although studies have shown that cement content did not greatly impact the shrinkage [31, 32], it is traditionally believed that excessive cement dosage can lead to material shrinkage and cracking.

2.4. Water. In CRME, before demulsification, water not only ensures the full adherence of emulsified asphalt to RAP materials during the mixing process but also has a lubricating effect on the compaction. Besides, part of the water undergoes hydration with cement, resulting in higher early strength. The total water content in the CRME not only includes the external water added but also the water contained in RAP materials and the emulsified asphalt.

3. CEAM and CRME

CEAM and CRME are two important materials in civil engineering. CEAM is mainly applied in the track engineering, and CRME is mainly applied in the pavement engineering. They have similarities and differences.

3.1. Similarities and Differences of Structure Functions. For CEAM, it is an essential component in the ballastless slab track system for high-speed railway (Figure 3) and its primary functions include supporting structures, adjusting the slab track's geometry during construction, and dissipating/reducing orbital vibration caused by the high-speed passing of the train, and transferring the train-passing stress to the



(a) (b)

Figure 3: Structure of a slab ballastless track; (a) China railway track system (CRTS) I, (b) CRTS II. CA mortar: CEAM [19].

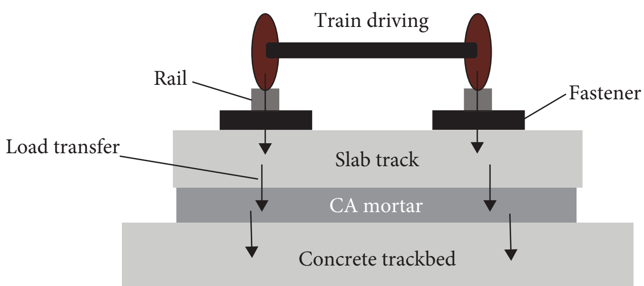


Figure 4: Schematic representation of layer arrangement and load transfer in the ballastless slab track system [19].

lower structure [33] (Figure 4). In addition, it can also be used as a sealing layer to prevent moisture intrusion into the subgrade and bedrock. Furthermore, its characteristics play a decisive role in the durability, safety, stability, comfort, and maintenance workload of slab ballastless track system [34–38].

For CRME, emulsified asphalt is applied as the binder and its primary function is to work together with RAP materials to form a whole and providing traffic capacity for roads or highways. Theoretically, CR technology can be applied in any pavement layer and in any volume traffic conditions, but due to some technological problems still ill-understood, especially in the process of design and construction [3], CR technology is commonly used in base/subbase course [39], and sometimes in binder course. For instance, in China the specification for the void content of CRME is 8%–13% [40]. However, that of HMA applied in surface course is 3%–6% [41]. The larger void content would make the asphalt prone to be oxidated and aged, resulting in loosing, peeling, cracking, etc. Besides, the precipitation could easily penetrate the structure, causing serious water damage to the asphalt pavement.

3.2. Similarities and Differences of Components. CEAM and CRME mainly consist of similar materials: emulsified asphalt, cement, aggregates, water, and other chemical additives.

Aggregates in CEAM are fine aggregates and mainly composed of sand [19], while aggregates in CRME are RAP materials and a small portion of mineral aggregates.

Stabilization agents of CEAM and CRME shared the same component, and both asphalt emulsion and cement were very important. For CRME, cement accounts for a very small proportion and acts as additive. However, it has played a significant role. Due to the effect of cement, the bonding ability between asphalt and RAP materials is greatly improved, and the strength growth rate of CRME is accelerated, shortening the time for CRME pavement to open to traffic [15, 42]. Traditionally, it was believed that with the increase of the amount of cement, the stabilized materials would become more brittle and less flexible. Although it could spread more stresses under the wheel loading, it would become more like cement-stabilized materials and give rise to shrinkage cracking, resulting in reflection cracking on the pavement surface. Although there was research indicating that increasing the cement content would not cause the abovementioned issues [31, 32], the specifications still limited the amount of cement used. For instance, in *Chinese Technical Specifications for Highway Asphalt Pavement Recycling* (JTG/T 5521-2019), it is advisable the cement dosage does not exceed 1.5%, and the amount of cement added in CRME should not exceed 1.8% [40].

For CEAM, the proportion of cement increases while the proportion of emulsified asphalt decreases. In engineering practice, there are mainly two types of CEAM. One with relatively low elastic modulus and strength employs a cationic asphalt emulsion, which is usually applied to a unit slab track. The other one with relatively high elastic modulus and strength is applied to the Bögl continuous slab track and usually employs anionic asphalt emulsion. The asphalt to cement ratio (A/C) is different between two types of CEAMs: the A/C ranges from 0.6 to 1.2 for the former, and 0.2 to 0.6 for the latter [19]. It is summarized in Table 1.

4. The Adhesion Development of Emulsified Asphalt Mastic

The adhesion development of CRME begins with the process of demulsification and hydration caused by the migration and dissipation of water inside the emulsified asphalt mastic. Currently, scholars worldwide have conducted extensive studies on this issue, and the study methods and important results are summarized in Table 2. (Since the stabilization agent of

Table 1: Characteristics of two types of CEAM.

		Type I	Type II
Application		CRTS I	CRTS II
Emulsifier		Cationic	Anionic
A/C ratio		0.6–1.2	0.2–0.6
Inject depth		50–70 mm	20–40 mm
Strength	28 days compressive strength	About 1.8 MPa	About 15 MPa
	Elastic modulus	100–300 MPa	7,000–10,000 MPa
Preferences		Requiring more damping performance	Needing more strength

CEAM and CRME are similar with each other, when searching literatures on the adhesion development of emulsified asphalt mastic of CRME, it is appropriate to simultaneously review the references of CEAM).

In Table 2, the research was roughly arranged in chronological order. It could be found that early studies evaluated the demulsification through measuring the stability of asphalt emulsion by electrophoretic testing. However, it lacked the ability to describe the microscopic behavior during the demulsification and hydration process. Within the past 10 years, study methods were gradually transitioning from macroscopic to mesoscopic and microscopic. Early research on the influencing factors of adhesion development of emulsified asphalt mostly focused on rock types (granite, basalt, and limestone [47]), while later research on influencing factors focused more on surface energy and chemical composition [55–58]. In addition, environmental scanning electron microscope (ESEM) and scanning electron microscope (SEM) were applied to observe the microstructure formation in early stage of adhesion development [26, 59–63].

5. Application of X-Ray Technology in Microscopic Study of CRME

X-ray computed tomography (CT) or X-ray microtomography (MT) is a nondestructive method for detecting the internal and external structures of objects. It is to record the spatial distribution of energy attenuation of X-rays when penetrating the detected object, and each volume element in the established 3D volume image represents the average attenuation of X-rays in the corresponding spatial part [64]. Therefore, for low-density areas, the image appears black, and as the density increases, the image appears whiter. CRME is composed of multiple raw materials, including solid phase, liquid phase, and gas phase. If X-ray CT or MT technology was applied to analyze the microstructure of CRME, the image of aggregates is the brightest, followed by that of emulsified asphalt mastic, and that of voids is the darkest. Hence, it has been widely used for the evaluation of the internal microstructure and performance of engineering materials [65].

Scholars worldwide have conducted relevant research on the void characteristics of CRME using X-ray CT, including the void content and the void distribution [66–68]. Some results of the air void content and the Weibull distribution modelare shown in Table 3.

From Table 3, this method could be used to determine the void content and the distribution of voids. Understanding the distribution pattern of internal voids was helpful for the mixture design and the improvement of the long-term performance.

CRME was composed of solid, liquid, and gas. The solid phase mainly refers to RAP materials and aggregates. The liquid phase mainly refers to the asphalt mastic. The gas phase mainly refers to voids. Except for the void content, scholars also study the distribution of asphalt using X-ray CT. The distribution of new and aged asphalt on the surface of RAP is shown in Figure 5 [69].

From Figure 5, the thickness distribution of emulsified asphalt was characterized. Understanding the distribution of asphalt is of great significance for improving the fusion of the new and aged asphalt, enhancing the adhesion of the fuzed asphalt, and optimizing the performance of the mixture.

In addition, X-ray CT technology could be utilized to analyze the fatigue crack development rule and cracking behavior [70]. It was meaningful for further understanding the mechanism of fatigue cracking for CRME and provided theoretical support for improving the fatigue cracking resistance.

In China, X-ray CT is mainly applied to the study of meso-structure, including geometric characteristics such as the aggregate form and morphology [71], the void information [72], and emulsified asphalt mastic properties [73]. The compaction method [74, 75], the curing method [76], and the additive dosage [77–79] could be optimized through this method. Further analysis of water sensitivity was also conducted to improve the performance of CRME [80, 81]. However, there were few ones on real-time tracking and simulation of its microstructure using this technology.

6. The Strength Development of CRME

The strength development of CRME mainly relied on the close adherence between emulsified asphalt mortar and RAP materials. In some research, the adhesion degree between emulsified asphalt mastic and RAP materials was studied through used quantitative evaluation [82, 83].

Water was the important intermediary for the development of adhesion in CRME, because of the release during demulsification and the absorption during hydration. Therefore,

Table 2: Important studies and results of research on the adhesion development of emulsified asphalt mastic.

References	Year	Study methods	Important results or explanations
[43–45]	2007–2009	Stern's double layer theory and electrophoresis experiments.	Detecting the stability of the asphalt mastic.
[46]	2015	Testing the time-varying law of the average particle size and observing it with an optical microscope.	Dividing the adhesion development into three stages: (1) The contact stage between cement and emulsified asphalt; (2) the dynamic equilibrium stage; and (3) the accelerated demulsification stage.
[47, 48]	2015	Studying the variation of the viscosity with time through the Gompertz model.	The Gompertz model could reflect the variation of adhesive viscosity over time.
[49]	2017	UV visible spectroscopy and Lambert Beer's law.	Introducing 2 parameters: UV absorbance and emulsifier concentration to characterize this process.
[50, 51]	2014–2018	Testing the effects of pH value and Ca^{2+} concentration through rheological shear tests and storage stability tests.	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.
[52–54]	2015–2022	Electrodynamics and centrifugal conductivity method.	Managing the content of Na+ and phenyl functional groups in the emulsifier could adjust the rate of demulsification process.
[26]	2018	Environmental scanning electron microscope (ESEM) and Scanning electron microscope (SEM).	Early-stage (<14 days) strength formation depends mainly on the integration between emulsion particles. Accelerating the demulsification by surfactant can improve the early-stage strength.
[55, 56]	2018–2019	Examining aggregate characteristics through ultraviolet spectrophotometry and the orthogonal test.	Influencing factors of demulsification speed: Chemical composition, surface energy, and specific surface area.
[57]	2020	Examined the demulsification time and influencing factors.	Increasing pH value: Delaying the cement hydration and prolonging the demulsification. Stabilizers: Shortening the demulsification. Increasing the stirring speed: Shortening the demulsification.
[58]	2022	Effects of the main chemical components of the aggregates through the Gompertz model.	Influencing factors of demulsification speed: Surface energy, specific surface area, and pH value of the chemical components.

Table 3: Results of air void content and regression parameters [67].

Compaction method	Air void content (%)			Weibull distribution model		
	Measured	CT image processing	Deviation	Scale parameter k	Shape parameter k	R-square
Static load	10.6	9.6	−1.0	1.818	1.815	0.998
Marshall	11.1	9.9	−1.2	2.580	2.160	0.999
Superpave gyratory compactor	11.5	10.3	−1.1	2.424	1.900	0.998

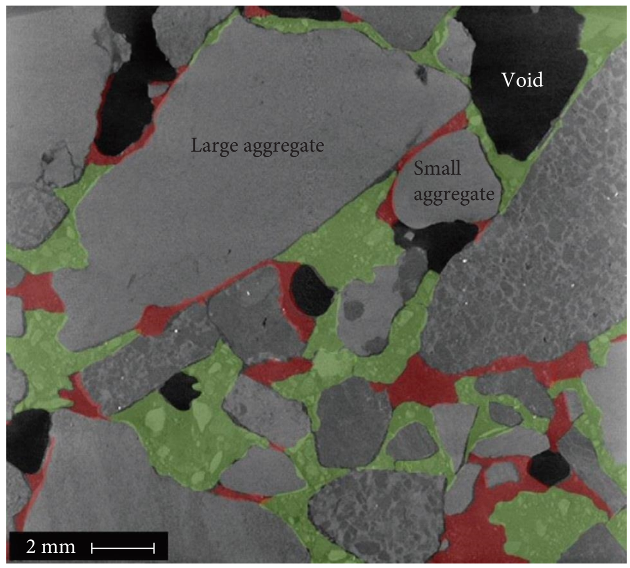


Figure 5: CT image with virgin binder highlighted in red and the RAP binder in green [69].

Table 4: Effects of influencing factors on CRME [14, 28, 62, 87, 91, 93].

Influencing factors		Effects
Curing temperature		The high temperature would accelerate curing.
Curing humidity		Cured in sealed conditions (high humidity) exhibited a slower curing rate and a higher long-term performance.
Materials	Cement dosage	Increasing it would increase the indirect tensile stress (ITS), the high-temperature stability and moisture susceptibility. CRME had better low-temperature cracking resistance with 1%–2% cement.
	Emulsified asphalt dosage	Increasing it would increase the resistance against rutting and fatigue.
	Water content	The right amount of water is conducive to the compaction.
	Aging degree of aged asphalt of RAP	It has a negative effect on the strength formation.
Compaction		The higher compaction power is beneficial to strength formation.

analyzing water loss and linking it with the mechanical properties of materials could be used to evaluate the strength development of CRME [84–86].

In addition, the absorption and release of water were completed in curing period. Curing was the process of placing CRME under specific temperature and humidity conditions to allow the demulsification and cement hydration inside emulsified asphalt mastic and develop the material strength. Therefore, many scholars focused their research on curing period and establishing a prediction model for material strength [87–92]. There were many influencing factors of CRME, including temperature, humidity, materials, etc. The effects of these influencing factors are summarized in Table 4.

Through the analysis of the adhesion development of CRME, it could be concluded that this process relies on water migrating and dissipating and it is the result of the fusion and interaction of new emulsified asphalt, cement, and aged asphalt on RAP surface. However, at present, there are still some problems in the research on this process. First, it could only point out the contribution of favorable univariate factors to adhesion growth, and it cannot measure the coupling effects of complex condition on the promotion of adhesion development. Second, it lacked a prediction model of the strength development that took curing temperature, humidity, and other influencing factor as variables, resulting in the limitation of the long-term performance improvement of CRAM.

7. Conclusions

In order to further promote the application of CRME, and better follow-up the research progress of CR technology, this paper first reviewed raw materials; second illustrated the similarities and differences between CEAM and CRME, and third discussed adhesion development of emulsified asphalt, microstructure, and the strength development of CRME. Conclusions can be drawn as follows:

(1) Raw materials of CRME are emulsified asphalt, RAP materials, cement, water, and other additives. Traditionally, it was believed that RAP materials could be considered as "black rock," and cement content would greatly impact the shrinkage. However, it was suggested by some studies that it was the opposite.

(2) CEAM and CRME mainly consist of similar raw materials: emulsified asphalt, cement, aggregates, and water. However, there are significant difference between each other in structure functions and proportion of each component.

(3) The adhesion development of CRME begins with the process of demulsification and hydration caused by the migration and dissipation of water inside the emulsified asphalt mastic. Early studies evaluated this process usually through macroscopic tests and within the past 10 years, study methods were gradually changed to mesoscopic and microscopic methods.

(4) X-ray CT or MT has been widely used to analyze voids characteristics and the distribution of asphalt in CRME. However, there were few ones on real-time tracking and simulation of its microstructure using this technology.

(5) The strength development of CRME used water as a medium. The migration and dissipation of water in the curing period was very important. Influencing factors of this process was studied thoroughly.

(6) The strength of cold recycled mixtures mainly relies on the bonding between stable materials and RAP. Conducting research on it will help clarify the influence of various factors on the strength development of cold recycled mixtures. It is of great engineering significance to improve the long-term performance of mixtures, extend their service life, and expand their application scope.

8. Suggestions for Future Research

Based on the above research, although researchers have conducted extensive research on the microstructure in adhesion development process and strength formation mechanism in early stage of CRME, there are still the following problems:

(1) The microbehavior mechanism of internal water transport in emulsified asphalt mortar has not yet been thoroughly revealed. Although researchers have been attempting to quantitatively characterize the process of demulsification and hydration, the results have only stayed at the macroanalysis stage and have not yet analyzed the transient changes in water transport and material morphology within the adhesive at the microscales and mesoscales, especially at the microlevel. X-ray technology may be helpful in this research.

(2) It lacked scientific measurement research on the promoting effect of complex conditions on the development of adhesive properties of mixtures. Most research adopts orthogonal experiments to analyze the impact of a certain variable factor on adhesion development. It lacked comprehensive coordination of the coupling effects of various conditions on the adhesion of the mixture, resulting in a lack of theoretical and scientific basis for predicting and improving the long-term performance of CRME.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

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