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## Aggregate pre-coating approach using rubber- and silane-coupled thermoset polymer and emulsion for warm-mix asphalt mixtures



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#### HIGHLIGHTS

• The modified epoxy coating film maintained above 90% after boiling for six hours.

• The adhesive strength of the modified epoxy resulted in seven times higher value.

• The modified epoxy could be used as an emulsified warm-mix asphalt mixture.

#### ARTICLE INFO

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#### ABSTRACT

This study proposes an aggregate pre-coating approach using both a synthesized thermoset polymer and an asphalt emulsion as a warm-mix asphalt mixture. An oil-in-water emulsified asphalt is added to initiate a moisture hardening reaction for aggregate coating with the polymer on the surface of the aggregate. Then, the PG64-22 asphalt binder is finally mixed with the pre-coated aggregates in the temperature range of 120–130 °C. The chemically modified epoxy can react with some polar components on the surface of aggregates, and it bonds with nonpolar groups in an asphalt binder so that it gives high adhesive strength on the surface of aggregates.

The surface strength of the synthesized epoxy was at least 2-fold greater than the general epoxy sample, as verified by the Dolly test. The chemically modified epoxy was used as a coating agent for aggregates and made possible to react with the water molecules in the emulsified asphalt binder. The bonding effect was verified by the boiling test, maintaining greater than 90% of the coated film.

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#### 1. Introduction

Asphalt is a thermoplastic material, which either exists in nature or is a by-product of petroleum distillation. Asphalt is made of various hydrocarbons and heterocyclic components containing nitrogen, oxygen, sulfur, and metal atoms. Although asphalt has been widely used as a binding material for a flexible pavement, due to the chemical complexity of the asphalt, it is impossible to characterize its chemical composition precisely [1].

The chemical composition of asphalt binder is highly dependent on the source of crude oil and a refining process used, and asphaltenes and maltens are the principal components of asphalt binder. The cohesive or adhesive strength of asphalt binder is strongly dependent on the partial fraction of saturates, aromatics, resins, and asphaltenes. Asphaltenes have the highest polarity,

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and polar fractions, such as asphaltenes or polar aromatic rings highly affects to the stiffness of asphalt binder [2].

Regarding the adhesiveness of asphalt binder on the surface of aggregate, although the polar fractions of asphalt binder may contribute to enhance the adhesive strength at the surface of aggregate, the weight fractions of asphaltenes and polar aromatic rings are about less than 15% and 20% of asphalt binder, respectively. The polar fraction of asphalt binder less than around 35% may engage to drive the adhesive strength on the surface of aggregate. The relatively small percent of the polar fractions contributing to the surface adhesive bond strength may cause early-age stripping or raveling failures [3].

In the context of the cohesiveness of asphalt binder, since a vacuum distillation unit (VDU) was advanced in the 21st century, refinery operations have started to increase the production of fuel oil from crude oil. In other words, causing the high extraction rate of the oil, the increase of physical (e.g., viscosity) and engineering (e.g., complex modulus) properties of asphalt binder may result from the relatively high polarity due to the high rate of asphaltene

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fraction. The polar components contributed to the elastic part while the nonpolar parts supported to the viscous part of the viscoelastic response of asphalt binder [4].

Therefore, the increased stiffness of asphalt binder due to the high fraction of polar components (i.e., asphaltenes) may require a chemical modification of asphalt binder to make it more ductile especially at a low temperature. Besides, to expect a higher viscosity at a high temperature, it needs to engage a polymerization process with an additional polymer, such as SBS (styrene-butadiene-styrene) or SBR (styrene-butadiene-rubber) [2,5].

SBS has been widely used to modify bitumen because of its excellent engineering properties. However, it is worth noting that the cohesive strength of a polymer-modified asphalt binder is enhanced by the increased cross-linking network of the tri-block copolymer of SBS in a short time though, some unsaturated midblocks may cause undesirable oxidative reactions of the carbon double bonds (C=C) in the styrene or polybutadiene segment. This reaction causes the degradation of the SBS modifier and the oxidation of bitumen itself so that the softening point of the modified binder results in an almost equal temperature to that of unmodified bitumen with less than 500 h of aging time in a pressure aging vessel [5,6].

The stiffening of the SBS dispersed in an asphalt matrix and the disappearance of cross-linked networks within a short period less than about one year of natural weathering need to be addressed due to the existence of carbon double bonds in the SBS which is more prone to oxidative aging [5,7–9].

Besides, ultraviolet light has enough energy to break the carbon double bonds in the elastomeric phase of the SBS, splitting the polymeric network into shorter chains, which promotes oxidation and makes the polymer more brittle and lose its elasticity gradually [10].

The relatively high energy consumptions during the chemical polymerization, drying aggregates, and mixing stages compared to the general hot-mix asphalt mixture and the phase separation of unsaturated polymers dispersed in an asphalt matrix phase due to long-term storage may be needed to be solved or to be substituted with a non-polymerized warm-mix asphalt mixture.

The insufficient adhesive or cohesive strength of asphalt binder may require additional functional molecular groups not only for making a greater density of cross-linking network through polymerization than the general asphalt binder has but also for the enhanced surface bond strength on the inorganic surface of aggregate. This study proposes an aggregate pre-coating approach with a rubber- and silane-coupled epoxy and an emulsified asphalt binder under a mixing temperature in the range of 120–130 °C to overcome those chemical and practical shortcomings regarding the adhesiveness and the cohesiveness strength at the interface between asphalt binder and aggregate.

#### 2. Selection of materials

#### 2.1. Rubber modification of thermoset polymer

The incompatibility of general epoxy resin and asphalt binder is the significant restriction in the use of epoxy resin as an aggregate pre-coating agent; although a previous work has confirmed that the epoxy resin can enhance the performance of asphalt binder. The incompatibility comes from the immiscible characteristics between a polar epoxide group and the non-polar components of asphalt binder [1].

This study proposes an engineered thermoset epoxy not as the polymer modification of asphalt binder but as the coating agent for aggregates as a pre-mixing process in a plant. A thermoset epoxy resin is synthesized under the relatively low temperature around 170 °C than the polymerization does to be as a surface coating agent of aggregates in this study.

The irreversible feature of thermoset polymer makes hard to use it as a paving material because of the difficulties in controlling a curing time and temperature. Because of the irreversible characteristic of the unmodified epoxy after curing and setting, the mechanical behavior of epoxy is usually represented by the brittle failure mode due to its high toughness even at a room temperature.

The brittleness makes the epoxy need to be more ductile to preclude the sudden failure; the epoxy resin can be more flexible by adding some rubber molecules to the epoxy resin.

The molecular structure of a diglycidyl-ether-bisphenol-A (DGEBA) epoxy resin is shown in Fig. 1 without modification.

Fig. 1 represents the general thermoset resin, which sets through an irreversible chemical cross-linking reaction, referred to as a curing initiated by an amine or isocyanate hardener. The curing makes the molecular structure of the epoxy rigid; brittle failure can occur when subjected to impact loading. Therefore, adding a rubber component, such as a carboxyl (–COOH)-termina ted-butadiene acrylonitrile rubber (CTBN) improves the flexibility of the epoxy, that is designed to react with the epoxide group of epoxy resin before the hardening reaction has proceeded [10–12].

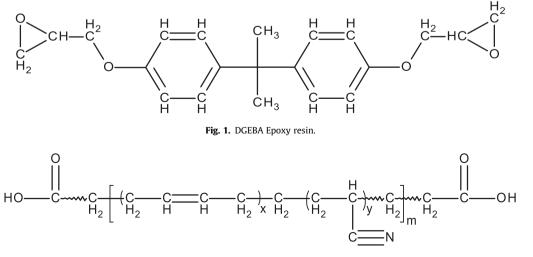


Fig. 2. Molecular structure of CTBN.

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