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Effect of chemical composition on rheology and mechanical properties of asphalt binder



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HIGHLIGHTS

- Derivative binders were developed by spiking the amount of individual SARA fractions.
- Polar fractions increase both stiffness and tensile strength of asphalt binder.
- Polar fractions increase stiffness at low frequencies but not as much at high frequencies.
- Polar fractions produce smaller and more cavities that nucleate tensile failure.

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ABSTRACT

Asphalt binder imparts most of its characteristics to the asphalt concrete mixture and also dictates several forms of distresses in asphalt mixtures and pavements. The chemical makeup of the asphalt binder dictates its rheological and mechanical properties. The objective of this study was to investigate the effect of the relative concentration of different polar fractions on the rheology and tensile strength of asphalt binders. Two different binders were separated into four (SARA) fractions based on their polarity. The fractions were individually doped in the original binder to produce four derivative binders from each of the two parent binders. Dynamic shear moduli and tensile strengths of the asphalt binders were measured using a dynamic a shear rheometer (DSR) and a tension-compression machine, respectively. The results show that an increase in the concentration of polar fractions resulted in an increase in both the stiffness and tensile strength of the binder. Addition of more polar fractions resulted in a substantial increase in stiffness at lower frequencies of loading but this increase was not as substantial at higher frequencies of loading. Observation of the failure surfaces from the poker-chip test also revealed that binders with higher polar fractions that had higher stiffness and tensile strength also had higher number and smaller size cavities that resulted in crack nucleation and failure. The findings from this study can help better understand the behavior of aged and chemically modified asphalt binders and also engineer better performing binders for pavement applications.

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1. Introduction and background

Asphalt binder, also known as bitumen, is a residue obtained from the distillation of crude oil and an important component of asphalt concrete mixture. The mechanical properties of asphalt binder are dependent on the time/rate of loading, temperature and age. Asphalt binder imparts most of its characteristics to the asphalt concrete mixture and also dictates several forms of distresses in asphalt mixtures and pavements. Hence, it is very important (but not sufficient) to ensure that asphalt binders used in the

production of asphalt mixes are inherently resistant to cracking, rutting and other distresses that a pavement may undergo. Binders with inherently superior performance characteristics or durability can be produced by one or more means depending on the economics of production (e.g. blending binders from different crude stocks, using specific refining processes such as, air blowing, and using chemical or polymer modifiers). In addition to asphalt binder production, binders may also be modified using chemical or polymer additives during the production of asphalt mixtures with or without reclaimed asphalt pavements (e.g. use of chemical modifiers or rejuvenators). Improving the durability of asphalt binders during refining or using post-production techniques and additives requires a better understanding of the relationship between the chemical makeup of an asphalt binder and its performance.

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Asphalt binders have a very complex chemical composition. Asphalt binders mostly comprise carbon (82-88%) and hydrogen atoms (8-11%) and one or more heteroatoms of nitrogen, sulfur, and oxygen; the heteroatoms, although small in percentage, are important because they impart polarity to the asphalt binders [15,27,30,34]. Moreover, the chemical composition of any particular asphalt binder also depends on the source of the crude oil and refining process used [13]. Due to the complex and diverse structures of the individual molecules that make up an asphalt binder, it is not feasible to model the exact chemical composition of the binder and its relationship with engineering properties. A more practical approach to understand the relation between chemical makeup and engineering properties of asphalt binders is to classify the chemical composition of asphalt binder based on different attributes rather than to identify the exact structure of all the molecules in any given binder. The commonly used attributes are molecular size, ionic character (acid, base, amphoteric) and polarity. Of these attributes, polarity of asphalt molecules has been found to be relatively more useful in understanding the relationship between binder chemistry and rheology [15,28,33,34].

The polar fractions in any given asphalt binder are typically classified into two groups, asphaltenes and maltenes. Maltenes are further separated by elution–adsorption liquid chromatography into saturates, aromatics, and resins [4,6]. The separated fractions (\underline{S} aturates, \underline{A} romatics, \underline{R} esins, and \underline{A} sphaltenes) are often referred to by their acronym SARA. Saturates have the least polarity and asphaltenes have the highest polarity; the polarity of aromatics and resins lie in between these two extremes.

The relationship between polar fractions of different asphalt binders to their physical (e.g. viscosity) and engineering properties (e.g. complex modulus) have been extensively studied by several researchers over the past five decades. For example, Corbett [6] demonstrated that saturates and aromatics impart soft or plasticizing properties, whereas resins and asphaltenes contribute to the stiffness of asphalt. Dealy [8] studied rheological properties of oil sand bitumen by changing the amount of asphaltenes and maltenes. His results showed a significant increase in viscosity of the oil sand bitumen with the addition of asphaltenes and a reduction in viscosity with the addition of maltenes. Robertson [31] studied the chemical composition of asphalt binder at the molecular level. He reported that polarity was the most important indicator of asphalt properties; he found that the polar molecules contributed to the elastic part of the binders' viscoelastic response while the non-polar contributed to the viscous part of the viscoelastic response. More recently, Robertson et al. [32] conducted an elaborate investigation of the relationship between binder chemistry and its physical properties. They reported that the viscosity of asphalt binder was significantly affected by the source and relative amounts of asphaltenes and maltenes. They reported that molecules with relatively high polarity (e.g. asphaltenes) would tend to associate more strongly due to the secondary intermolecular forces of attraction. Such association would result in resistance to shear stresses when external loads are applied. In other words, the presence and association between such polar molecules were regarded as the viscosity or stiffness building components. As empirical evidence, similar to Dealy [8], they also reported that in comparison to a reference binder, the binder with a higher amount of asphaltenes showed much higher viscosity, and the binder with a higher amount of maltenes showed less viscosity.

Michalica et al. [24] investigated chemical and physical properties of two asphalts from different sources (Urals, Russia and Cold Lake, Canada). Besides the amount of chemical components, they identified other factors, such as "chemical structure, polarity, molecular weight, and their mutual interaction" that control the asphalts' behavior. They found that the binder with lower asphaltenes content (Ural asphalt) was more stiff and brittle at lower

temperature than the binder with higher asphaltenes content asphalt (Lake asphalt). The results of Michalica et al. [24] combined with the results from other researchers present an interesting finding. First, it appears that the stiffness of the binder cannot be globally related to merely the asphaltenes or maltenes content because these fractions may have different properties depending on the source of the binder. Also, it appears that in some cases, relatively higher asphaltenes content does not necessarily imply increased stiffness or toughness (particularly at low temperatures). In fact, similar findings will be demonstrated in the latter part of this study.

Some researchers have tried to investigate the micro structure of different molecular species or morphology of the asphalt binder as a means to better understand the relationship between binder chemical composition and its engineering properties. Loeber et al. [19] studied asphalt microstructure using scanning electron microscopy (SEM) and atomic force microscopy (AFM). They were perhaps amongst the first researchers to report a dispersed phase in the microstructure called the "bee" structure. They assessed that the presence of asphaltenes is responsible for this kind of structure. Pauli et al. [25] also investigated the relationship between asphalt microstructure and chemical composition. They proposed that the "bee" structures were present in asphalt binders with high asphaltenes content. In subsequent work, Pauli et al. [26] hypothesized that the structures were due to the presence of waxes or aliphatic chains in the asphalt binders. The "bee" structures along with other material phases were observed by Masson et al. [22], Masson et al. [23] and Jäger et al. [14]. Masson et al. used phase detection microscopy (PDM) to observe asphalt microstructure and defined material phases as "catana", "peri", "para" and "sal". They suggested that catana phase represented bee structure (asphaltenes) which was surrounded by periphase (resins and aromatics); periphase was further surrounded by paraphrase (saturates). They also observed each para phase contained sal phase, which was of the lowest molecularweight amorphous alkanes. Allen et al. [2] studied the micro-rheology of these phases using AFM before and after oxidative aging of different binders. Allen et al. [1] evaluated these phases for asphalt binders with varying concentrations of the different polar fractions. They observed that oxidative aging increased the area of catana phase. This finding was consistent with the results from previous studies [16,27]. Allen et al. [1] also reported that the concentration of different polar fractions had a significant influence on the size, distribution and micro-rheology of the different phases.

In summary, previous studies strongly support the idea that the amount and characteristics of chemical fractions significantly affect the rheological and mechanical properties of asphalt binder; however, to date there has been limited work done to understand the influence of these fractions on the mechanical properties of asphalt binder. In particular, there are very limited studies that quantify the influence of individual polar fractions on the properties of binder that are directly related to distresses (e.g. tensile strength).

The objective of this study was to investigate the effects of chemical composition based on SARA fractions on the rheology and tensile strength of a specially designed set of asphalt binders. Section 2 of this paper presents the materials and experimental testing that were used to achieve the objectives of this study. Section 3 presents the results and discussion; conclusions are presented in Section 4.

2. Materials and experimental testing

2.1. Materials

2.1.1. SARA fractionation

The main objective of this study was to systematically and quantitatively evaluate the influence of chemical fractions (based on polarity) on the mechanical properties of the binder. To achieve this, it was necessary to select or produce a set of

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