

Estimating Blending Level of Fresh and RAP Binders in Recycled Hot Mix Asphalt

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Executive Summary

Despite all advantages of using Recycled Asphalt Pavement (RAP) in new HMA mixtures, the lack of a clear understanding of the nature, and more importantly, the rate of blending between fresh and RAP material has been a major concern. Although it is expected that the degree of blending between the RAP and fresh binder is dependent on mixture temperature, contact time, and constituent binder chemistry, complete or near complete blending is often assumed in order to achieve the full cost savings potential of using RAP. Inadequate performance of some high-RAP mixtures has prompted several recent studies to call into question the degree of blending that truly occurs in high-RAP mixtures.

Since the aged binder exists as a thin layer covering the aggregates in RAP material, the mechanical blending through mixing between fresh and RAP binder during mixing is not a sound assumption. A better assumption is that blending between fresh and aged binders in contact is achieved through molecular diffusion driven by molecular energy imbalance and polar attractions.

This research presents an experimental-based method to quantify the time and temperature sensitivity of the diffusion rate and ultimate degree of blending that occurs between highly aged and fresh binders. Thin binder samples were conditioned at predetermined temperatures and time intervals to inspect the effect of these factors on the diffusion process between two binders. The rheological response of the binder samples were determined using the Dynamic Shear Rheometer (DSR), and was used in an analysis procedure developed to estimate the diffusion rate and blending capacity. Theoretical calculations for composite material were used to verify the diffusion rates and degree of blending estimated from laboratory measurements. The effect of

binder compatibility was investigated using two binder sources with extreme differences in composition. In-service blending capacity was also demonstrated for two different RAP materials.

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1 Introduction

1.1 Background

In recent years the world prices of crude oil increased significantly affecting the produced asphalt binder. Considering the point that most of produced asphalt binder is used in highway industry to construct new pavements, it is necessary to mitigate some of the expenses. Despite of benefits to solve environmental issues related to RAP, reclaimed material due to its cheap price and availability became very favorable in pavement industry. The limited natural source of granular material used in the asphalt mixtures is another reason to use RAP in the new pavements.

The first pavement containing RAP was implemented early 1970s in state of Nevada. In short time the usage of RAP in pavements became a favorable practice for pavement industry accepted by most of states (Federal Highway Administration, 2010). Since then, an ascending trend of producing and using RAP has been seen in asphalt pavement industry in United States.

According to National Asphalt Pavement Association (NAPA) currently approximately 500 million tons of new asphalt pavement material is produced in U.S., and this amount includes 60 million tons of reclaimed material reused or recycled into pavements. This indicates an increase of 72 million tons of RAP used annually comparing to early 1990s.

Several State of the Practices has been developed by federal government or by the state DOTs. A survey performed by Federal Highway Administration showed that, in most states the use of RAP is not mandatory and depends on economic considerations and availability of material. Another investigation by North Carolina Department of Transportation (NCDOT) in

2007 indicated that all 50 states allow for using RAP in HMA mixtures. The national average usage of RAP in the same year was estimated to be 12 percent, increasing to 15 percent by 2010 (West 2010). However just few states allowed for low amount of RAP in the asphalt wearing course because of their performance concerns. As it is shown in the Figure 1, 31 states allow using up to 19% RAP in surface layer while just 3 states has specifications for using up to 29% RAP in the HMA surface layer.

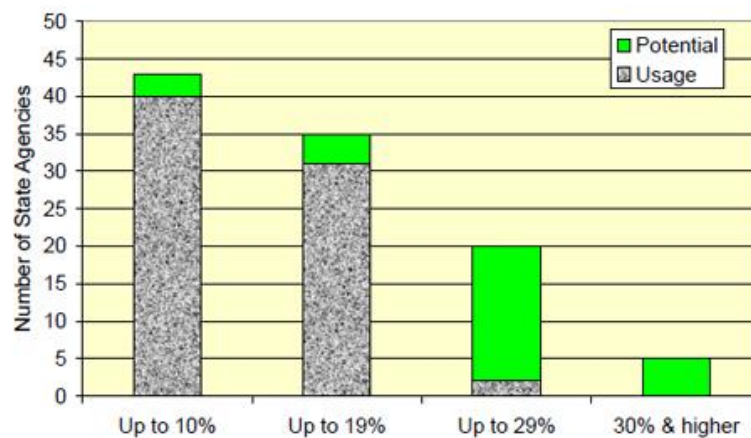


Figure 1 States allowable usage of RAP in the surface layer (Federal Highway Administration, 2011)

Despite of the point that by 2011 more than 40 states allowed using over 30 percent RAP in asphalt mixtures, just 11 states reported application of more than 25 percent RAP in practice (Nash, Sholar and Musselman 2011).

The current State of the Practice for using RAP in HMA pavement (AASHTO M323) presents some specifications. The criteria for selecting fresh asphalt binder depend on percent of RAP used. For high percentages of RAP (more than 25%), a linear blending chart should be developed which gives the target specifications to select the fresh binder performance grade (PG). This Practice assumes full blending between fresh and RAP binder. However based on a number of

studies (e.g. NCHRP 9-12 and Carpenter, et al., 1980) a full blending assumption may not be very reasonable. On the other hand the extracted RAP binder used for this procedure may bring up some concerns about the reliability of results. Based on previous studies there is a large variability in physical properties of extracted binders along with its difficulty, cost and hazardous effects (Stroup-Gardiner & Nelson, 2000, Kondrath 2009, Peterson et al. 2000).

Regarding these concerns, a procedure to estimate the amount of blending between fresh and RAP binders which there is no need for extracting binder, may be more favorable. The procedure should be able to determine the effect of exposure time and temperature for two binders in addition to the effect of different RAP and binder sources.

1.2 Problem statement and Hypothesis

The use of RAP and RAS materials in the asphalt industry is commonplace; however, many concerns exist regarding performance implications of pavements containing a high percentage of these materials. There are different points of view on whether the reclaimed binder material should be considered as 'black rock', in which no effective recycled binder is considered in design, or to consider binder coating RAP capable of replacing a portion of added binder by blending with fresh binder, thus potentially significantly effecting mixture properties. The NCHRP 9-12 study concluded that reclaimed asphalt material (RAP and RAS) does not behave just as black rock. Therefore, the performance of high RAP and RAS mixtures is dependent on the blending that occurs between the reclaimed and fresh asphalt binders. Failure to account for the blended binder properties can result in premature pavement distress.

Therefore, if it is considered that the blending between fresh and RAP binders is mostly controlled by mixing time and temperature, proper criteria for mixing will be beneficial for producing RAP mixes in the field.

The blending between two binders during mixing, as well as eventually during service life, is due to the molecular diffusion process, driven toward the establishment of a stable thermodynamic equilibrium state (minimum energy state). It is assumed that the blending between fresh and heavily-aged asphalt binder occurs as a result of two mechanisms: arbitrary motion of molecules at the interface, and mass transfer due to different concentrations of the two constituent materials.

Thus the hypothesis of this research can be summarized as follows:

- The blending between two binders during mixing and pavement service life of pavement can be estimated using molecular diffusion models.
- The time-temperature dependency of the blending can be modeled using diffusion relations based on the known rheological properties of the aged and fresh binder and the film thickness.
- The DSR can be used to provide an approximation of effect of blending of two binders on the rheology of the blended binder. It is expected that the G^* of the blended binders (as an indication of blending) will increase at a high rate at the onset of conditioning and but at a lower rate with increased conditioning time.
- The procedure of testing RAP mortars can be used in practice to estimate the blending between fresh and RAP binder at the intended production conditions for temperature and time.

1.3 Research Objectives

The blending between fresh and heavily aged asphalt binder is assumed to be controlled by diffusion during mixing. The diffusion rate is highly dependent on their viscosity and conditioning time. The DSR can be used to provide an approximation of effect of time and temperature on the rheology of the blended binder. After accounting for the effect of oxidative aging due to high temperature conditioning, a relationship between mixing time and temperature can be achieved for a combination of a fresh and aged binder. The main objectives of this research are:

1. Application of a DSR based procedure to measure the blending that occurs between two binder samples considering conditioning time and temperature.
2. Verify the linearity of simple blending charts (analogous to AASHTO M323) for heavily aged asphalt binder and fresh binder.
3. Verify that the model can be used for binders with extreme chemical compositions. Also, verify that DSR procedure can be applied for mortars produced from mixing fresh binders with RAP material containing aged binders.

2 Literature Review

2.1 Motivation

Application of RAP materials in new implemented pavements has gained a lot of interest due to the rise in crude oil prices during the last few years, limited aggregate resources and also for the sake of sustainability. Using recycled material in asphalt concrete is advantageous to preserve natural and energy resources (Bonaquist 2011). Studies in the past revealed a performance comparable to non-RAP HMA (Wiser 2011, Copeland 2011, Hong 2010).

Using RAP is preferred over the fresh material considering the increasing the cost of asphalt, shortage of aggregate sources and environmental considerations and lower cost comparing to non-RAP HMA mixes (Page and Murphy 1987). Producing HMA pavements containing 20 to 50% RAP may reduce the initial cost of new asphalt mixtures up to 14 to 34% relative to mixtures containing no RAP (Kandhal and Mallick 1997). Another benefit of using RAP is to reduce the waste produced and provide a good solution for disposal problems of highway construction materials (Al-Qadi, Elseifi and Carpenter, Reclaimed Asphalt Pavement - A literature review 2007). In 1996, about 33% of asphalt pavement in United States was recycled and reused in HMA (Sullivan 1996) . After a while the performance of asphalt pavements brings need to be replaced or rehabilitated. The unusable HMA layer is milled or excavated, crushed and screened to satisfy the gradation of the new designed HMA layer. The recycled material is added to aggregates during the HMA production procedure. As long as RAP material is available and much cheaper than fresh material, application of higher contents in HMA mixture is more favorable. Higher content of RAP used in the HMA will improve its Rutting susceptibility while initiates some other issues such as low temperature and fatigue performance. Considering a full

blending between two binders as a base assumption in AASHTO M323, should be evaluated using a procedure without binder extraction. Comparison the behavior of extracted binder may be questionable comparing to RAP binder covering aggregates which is not available for a full blending like the procedure used in the design guide. Therefore a procedure that can determine the interaction between fresh and RAP binder during mixing and while they are in contact at high temperatures, may provide a better understanding of blending level. This research presents a limited study of interaction between aged and fresh binder using performance properties.

2.2 Asphalt binder

2.2.1 Chemical structure of asphalt binder

As the physical properties (e.g. viscosity, stiffness etc.) are controlled by chemical constituents of asphalt binders, gain an understanding of its compositions is helpful for explaining the behavior they exhibit in blending. The composition highly depends on the crude oil sources; however it can be divided to four different matters:

Saturates

Colorless or light color liquid at room temperature, they usually form 5 to 15% of asphalt binder (Corbett 1969, D. Lesueur 2009). Saturates typically have the lowest molecular weight comparing to other fractions of asphalt binder (470-880 g/mol based on ASTM D-4124).

Aromatics

This component takes a high fraction of asphalt binder (30 to 45 wt.%). They have yellow to red color in room temperature (Corbett 1969). Their viscosity is higher than saturates at the room

temperature (D. Lesueur 2009). The molecular weight is 570 to 980 g/mol according to ASTM D-4124.

Resins

They form 30 to 45 wt.% of asphalt binder which in contrast with two former components, forms a black solid in room temperature (Corbett 1969). The molecular weight of resin is the highest after asphaltenes (780-1400 g/mol). These components disperse and stabilize asphaltene particles (called “asphaltene micelles”) in maltene phase. Maltenes includes resins, saturates and aromatic.

Asphaltenes

Asphaltenes usually form between 5 to 20% of asphalt binders and they are dark brown to black powder in room temperature (Corbett 1969) and are responsible for black color of asphalt binder (D. Lesueur 2009). These components are more studied because their role in building viscosity (Branthaver, et al. 1994). Their molecule weight is the highest among other constituents (800 to 3500 g/mol).

The first colloidal models for asphalt binder suggested by Rosinger in early 1910s (Rosinger 1914). In 1923, Nellensteyn and Errera presented a description of colloidal structure of asphalt binder (F. Nellensteyn 1923, Pfeiffer and Saal 1940). Nellensteyn speculated that asphaltenes form a colloidal suspension in the maltene phase (F. Nellensteyn 1924). Colloidal model was further developed by Pfeiffer and his colleagues to describe the rheological differences between what they called “sol” and “gel” asphalt binders (Pfeiffer and Saal 1940, I. R. Saal 1950). In their definition the sol binders showed a Newtonian behavior while the gel binders (mostly blown binders) were explained as non-Newtonian. They also suggested that most of asphalt binders fall

between these two extreme behaviors, called “sol-gel” binders. However in the modern definition, the non-Newtonian behavior of asphalt binder is described as “delayed elasticity with some non-linearity in viscoelastic properties” (R. Saal 1940, D. Lesueur 2009).

Modern colloidal model of asphalt binder

Before developing the modern model, the asphalt binder was described as colloidal formed of asphaltene micelles dispersed in maltene phase by resins (D. Lesueur 2009). Studies using Small Angle X-Ray Scattering (SAXS) and Small Angles Neutrons Scattering (SANS) approved that asphaltenes form micelles in organic solvents (Dwiggins 1965, Bardon, Barré, et al. 1996). Later studies on viscoelastic properties of asphalt binder, suggested that aforementioned micelles exist and they have Brownian motion at appropriately high temperatures (Lesueur, Gérard, et al. 1996, D. e. Lesueur 1997). The formation of micelles in the asphalt binder (sol or gel) and their Brownian motion defines the viscoelastic behavior of whole material. As the viscoelastic effects such as relaxation depends on the Brownian motion of micelles in the maltene phase (D. Lesueur 2009).

2.2.2 Aging of asphalt binder

Complex molecular organization and different phases, complicates the physical behavior of asphalt binder. In addition, chemical aging may cause further changing in the molecules organization through oxidation and polymerization (Read and Whiteoak 2003, Traxler 1961). Also lighter components of binder evaporate and a heavier, hardened material is the result (Read and Whiteoak 2003). This hardening effect may reduce the deformations under the compressive loading, but increase susceptibility to cracking. However the crude source is major factor for

chemical aging (D. Lesueur 2009). Produced asphalt binder may experience two other chemical oxidative aging during mixing and during the service life in HMA pavements.

The former, so called short term aging, occurs because of asphalt binder is in contact with oxygen a typically high temperature. This change in properties is simulated using the Rolling Thin Film Oven Test (RTFO) in the laboratory (D. Lesueur 2009). The viscosity after running this test is usually twice of the viscosity for fresh binder (Christensen and Anderson 1992). The asphaltene content increases by 1-4% of the total weight (Groupe National Bitume 1997).

The second type of aging, so called long term or in service aging, depends on the availability of oxygen around the binder (basically the position of binder in HMA layer) (D. Lesueur 2009). The Pressure Aging Vessel (PAV) developed by Bahia & Anderson (1995) is used to simulate the long term aging in the lab. The binder is kept in a pressure vessel at a relatively high temperature and exposed to oxygen at a high pressure for 20 hours. It is assumed that this procedure will accelerate the oxidative aging in laboratory giving change in properties after 7-10 years of service life. This test is desired to mimic the long term aging because (Bahia and Anderson 1995):

- It minimizes the volatilization
- There is no need of high temperature for aging
- Sample size that can be produced in this test is large enough
- It is practical in laboratory

Effect of aging on chemistry of asphalt binder

The aging changes some phases of asphalt binders to the others. Based on studies done on chemical compositions of asphalt before and after aging, fraction of aromatics decreases while

the resin content increases. Finally the resin phase converts to asphaltene gradually (D. Lesueur 2009, Wright 1965, Read and Whiteoak 2003). Saturate content remains the same due to its low chemical activity (Figure 2).

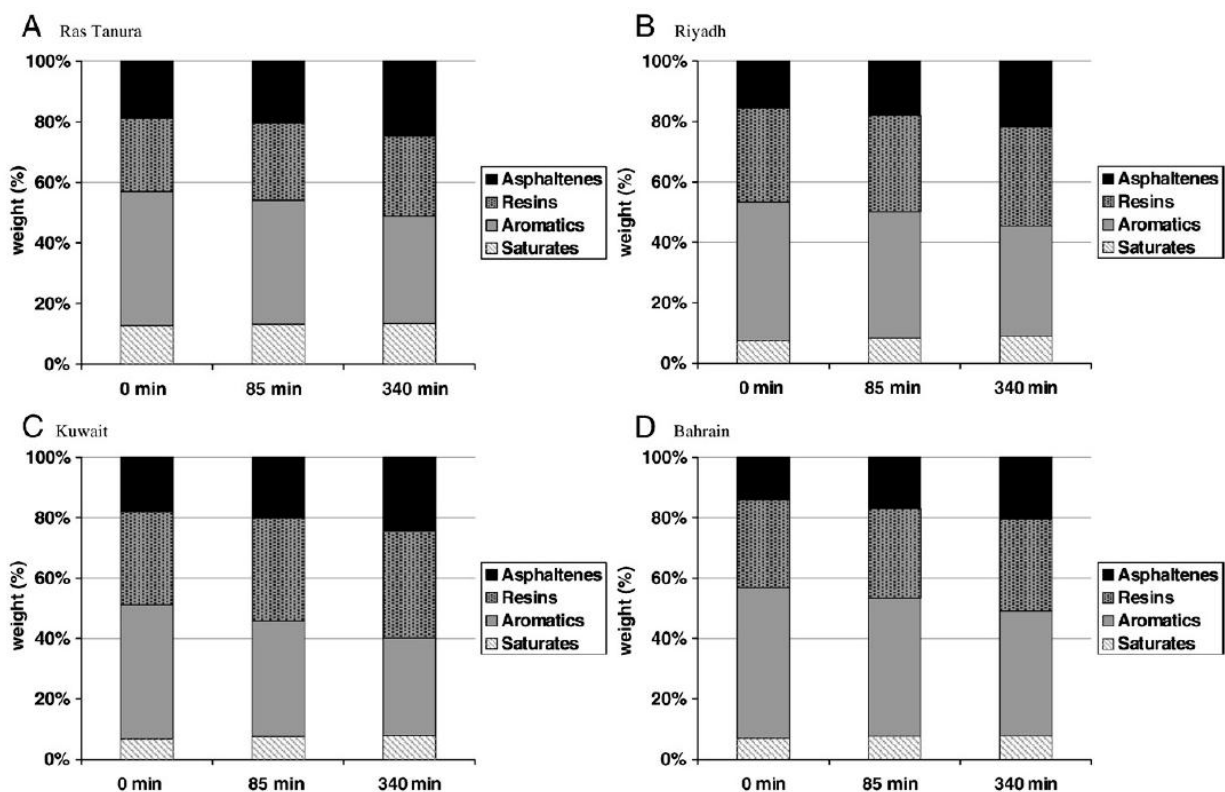


Figure 2 SARA analysis for different asphalt binders before and after 85 and 340 min RTFOT aging (after Farcas, 1998)

The newly produced asphaltene due to aging in the asphalt binder may not be the same as initial ones (Corbett 1969). In a study conducted by Groupe National Bitume asphalt binder from actual pavement were recovered and analyzed for a 90 months period. A linear increase was observed for asphaltene production and the field (between 2 to 10 wt.%) (Groupe National Bitume 1997). Aging of asphalt binder initially is linear then tends to slow down with progress of reaction. This is based on oxygen diffusion in the medium (Wright 1965, Petersen 1993).

Unaged and softer binder provides a lower viscosity and a higher diffusion rate of oxygen. Stiffened aged binder has higher viscosity and results in a lower amount of diffusing oxygen.

Considering high impact of aging on mechanical properties of asphalt binder, numerous studies has been conducted on its mechanism. All studies show change in rheological properties of asphalt binder due to aging. On the other hand the aging changes the balance between maltene phase and asphaltene fractions. This will affect the viscosity and concentration of asphaltene micelles in the asphalt binder confirmed with testing asphalt binder extracted from RAP material. the viscosity measured for recovered binder is significantly higher than unaged binder's. It is speculated that higher viscosity will inhibits the Brownian motion of asphaltene micelles and as result the interdiffusion. Therefore the level of aging in RAP asphalt binder may have a great impact on blending with fresh binder.

2.3 Current State-of-Practice: RAP Mixture Design and Other Applications

In 1970s, due to increased cost of crude oil, recycling asphalt pavement gained popularity in the pavement industry. First trial of using RAP in HMA was done more than 35 years ago in Texas and Nevada. Few years later State transportation departments started conducting the Demonstration Project 39 to provide a report on effectiveness of using RAP in new pavements to mitigate the expenses. The first Demonstration Project 39 was provided by Washington Department of Transportation in 1978. In 1980, National Cooperative Highway Research Program (NCHRP) published *Guidelines for Recycling Pavement Materials* (Copeland et al. 2011). Thereafter use of RAP seems not to be only an advantageous alternative for fresh material but also a qualifier necessity to ensure the competitiveness in flexible pavement industry (Al-Qadi, Elseifi and Carpenter, Reclaimed Asphalt Pavement - A literature review 2007). Based on

an FHWA and U.S Environmental Protection Agency report in early 1990s, more than 90 million tons of asphalt pavements were reclaimed every year which has increased in recent years. Most of these materials were recycled and used in HMA. RAP materials provide a source of aggregates and also substitute a part of fresh asphalt binder in new HMA. Reclaimed asphalt pavement material also can be used in sub-layers (base and sub-base), embankment or stabilized layer in the new pavements. HMA layer is constructed of high quality material, therefore RAP carries a very high quality as well (recycled from the same layer of pavement) and can replace expensive aggregates and binder in wearing course (Copeland et al. 2011).

Along with widespread application of RAP in pavement Asphalt Institute also developed some blending charts to simplify HMA design containing RAP which was in use by many states. Considering the durability problems of HMA with high content of RAP most of the states have limited the amount of RAP content within 10 to 50% (which high percentages are not used in practice).

After developing Superpave HMA design method some guidelines for application RAP were developed by Federal Highway Administration's RAP expert task (Bukowski 1997). These guidelines applied a tiered approach to determine the tests specifications used in design of HMA with RAP (Al-Qadi, Elseifi and Carpenter, Reclaimed Asphalt Pavement - A literature review 2007). NCHRP report 9-12 also validated these guidelines (McDaniel, et al. 2000). Kandhal and his coworkers recommended that using up to 15% of RAP does not require any considerations for changing asphalt binder performance grade (PG), however for higher contents of RAP up to 25%, one grade decrease in high and low performance temperature of binder. For RAP content above 25%, blending chart should be used to determine the allowable RAP content (Huang and Li 2005).

Level of changing properties of fresh binder due to interaction with RAP binder has always been a major concern (Al-Qadi, Elseifi and Carpenter, Reclaimed Asphalt Pavement - A literature review 2007). If RAP does not contribute in the HMA actively, the mixtures' properties will just depend on fresh binder. Generally it is believed that the RAP does not act just as "black rock" in the mixture and blending occurs between aged and fresh asphalt binders (Bukowski 1997). The RAP binder will induce some effects when is combined with the new binder changing the binder grade (McDaniel et al. 2000). Therefore it is expected that the aged binder will change the fresh binder properties.

NCHRP 9-12

This project was conducted to provide guidelines for applying RAP in Superpave system. Developing a manual that can be used in laboratory and pavement technicians dealing with RAP was another goal of the project. The effect of RAP on performance grade of binder, aggregates parameters, and mixture properties were considered as well.

During NCHRP project 9-12 HMA samples were provided to determine if there is interaction between fresh and aged binder in HMA containing RAP material. The superpave tests on HMA samples showed that in lower portions of RAP in the mix, there is no significant change in properties due to RAP binder, while in high contents, the properties noticeably changes. However for samples provided with blended binder (extracted binder with fresh binder) and normal RAP mixed with fresh material, no major difference were observed.

Phase two of the project merely considers the hardening effect of RAP aged binder on fresh binder. McDaniel et al. performed binder tests on the same RAP sources based on AASHTO

MP1. The results indicated the applicability of MP1 tests to RAP binders and linearity of the blending equation.

Phase three was performed to determine the effect of RAP on the mixture properties. The same sources of RAP and fresh binder were used in this phase. Shear and indirect tensile tests showed the stiffening effect of RAP binder at higher contents of RAP. Again the effect of RAP binder in lower contents was minor.

The report suggests that conventional Superpave binder tests can be applied to determine amount of allowable RAP used in the HMA or required number of performance grades changes for fresh binder based on a favorable amount of added RAP. The project also recommended that it may be unreasonable to assume that full blending occurs between fresh and RAP binder, but partial blending is very probable. Therefore the HMA with high RAP contents may increase the stiffness of fresh binder due to interaction with aged binder and this should be taken in consideration for selecting the fresh binder (changing PG or using blending chart). McDaniel et al. also recommended that the effect of RAP aggregates should be taken in consideration as well.

AASHTO M323

The current State of the Practice for using RAP in asphalt mixtures is based on AASHTO M323. This standard provides the mix design specification for HMA. The binder grade selection depends on the RAP content used in the mix (Table 1).

Table 1 Specification for selecting binder for RAP mixes (AASHTO M323)

Recommended fresh asphalt binder grade	RAP Percentage
No change in binder selection	<15
Select fresh binder one grade softer than normal	15 to 25
Follow recommendations from blending charts	>25

For RAP content higher than 25% a blending chart should be developed by determining physical properties of RAP binder. The RAP binder is recovered based on AASHTO T39 and tested in DSR to determine the critical high temperature. Then DSR test is performed on RTFO-aged recovered binder to obtain critical high temperature based on RTFO DSR specifications. Critical intermediate and low temperatures are figured out for RTFO-aged binder using DSR and Bending Beam Rheometer (BBR) respectively. Next step includes blending fresh RAP with known portions of recovered RAP. After determining the properties of blended binder, the required properties of fresh binder can be obtained.

This practice assumes a full blending between RAP and fresh binder which may not be obtained in the field.

2.4 Blending between fresh and aged binders

In practice, the RAP binder is a thin layer covering RAP aggregates and does not exist as a free mass in the mixture. Therefore it may be unreasonable to expect mechanical blending between RAP and fresh binder during mixing. The blending process can be explained using other phenomena which can help two binders blend when they are exposed at appropriate temperatures for a required time. This mechanism is called diffusion, which enables molecules transfer in the matter when they have enough energy to move.

Diffusion

The arbitrary motion of molecules (called Brownian motion) causes mass transfer in the medium. To initiate this transfer, the molecules should gain enough energy to move through the matter. This phenomenon known as diffusion can be affected by several factors such as shape

and size of molecules, temperature, forces between molecules, structural rigidity of diffusing molecules and etc. (Cussler 1997). According to Cussler, the diffusion rate has an inverse relationship with the viscosity of the fluid.

The diffusion of material is defined by diffusion coefficient indicating the mobility of material due to diffusion. Theoretically, the diffusion coefficient is calculated using first and second Fick's laws.

Fick's law for determining Diffusion coefficient

“Diffusion is the process by which matter is transported from one part of a system to another as a result of random molecular motion” (Crank 1975). Diffusion is process which an amount of a particular material transfers inside the other material due to Brownian motion of molecules. Diffusion coefficient is defined as amount of diffusing material across a unit area during 1 second,

Fick (1855) adjusted the same equations as used for heat conduction and put it in quantitative basis to calculate the diffusion. The basic assumption of the mathematical theory of diffusion is an isotropic substance; therefore, the rate of transfer of diffusing matter through the unit area is proportional to the perpendicular gradient of concentration measured in the section:

$$F = -D \frac{\partial C}{\partial x} \quad [1]$$

Where F is the rate of transfer per unit of area, C is the concentration of diffusing material, x is the space coordinate and D is diffusion coefficient. This equation is the basis for the differential equation of diffusion in an isotropic matter (Crank, 1975). If the diffusion coefficient is considered constant, the second Fick's law equation for diffusion is obtained as:

$$\frac{\partial C}{\partial t} = D \frac{\partial^2 C}{\partial x^2} \quad [2]$$

Where:

C: Concentration of diffusing material

t: Time

D: Diffusion coefficient [mm²/sec]

x: Vertical distance from the bottom DSR plate to the top of each sub-layer *n*, [mm]

These two equations (1 and 2) are known as Fick's first and second laws, respectively. As long as the material is assumed to be isotropic and homogeneous, Fick's second law can be used, but if not, the diffusion coefficient should be considered as a function of concentration change and another derivation of the first law should be used (Crank 1975).

Mathematical solutions proposed for Fick's laws, are usually based on the case of application and type of material. Crank (1975) derived solutions for constant diffusion coefficients based on initial and boundary conditions. The solution used in this research is derived based on assumption of finite system, cylindrical sample and using "method of separation of variables" explained as following:

$$C(x, t) = \frac{4C_0}{\pi} \sum_{n=0}^{\infty} \frac{1}{2n+1} e^{\left(-\frac{D(2n+1)^2 \pi^2 t}{l^2}\right)} \sin \frac{(2n+1)x\pi}{l} \quad [3]$$

Where:

*C*₀: Initial concentration of fresh binder, [%]

n: Sub-layer, [1, 2, 3, ..., 10]

t: conditioning time, [sec]

l: 'Artificial RAP' binder layer thickness, [1 mm]

x: Vertical distance from the bottom DSR plate to the top of each sub-layer *n*, [mm]

D: Diffusion coefficient, [mm²/sec]

This equation will be further explained in mythology section. Limited studies have been done on diffusion mechanism between fresh and RAP binder. Karlsson and Isacsson started a series of investigations of this mechanism in asphalt binder in 2000 briefly described as bellow.

Karlsson and Isacsson

Karlsson and Isacsson conducted a comprehensive study on diffusion of rejuvenators into asphalt binders. Their experiments were performed using two different procedures. First they used Fourier transform infrared spectroscopy by attenuated total reflectance (FTIR-ATR) to determine the effect of aging on the diffusion process (Karlsson and Isacsson 2002). They used six binder types with different penetration grades. One of the binders was aged in the RTFO for a specified time to reduce the penetration grade from 180 to 65. A relationship was developed between penetration and aging time. The diffusion was monitored using one commercial rejuvenator. They also used lauric acid and diphenylsilane as markers in the study. Thin layers of compound material were placed on a zinc plate and diffusion was monitored by absorbing the infrared spectra in FTIR-ATR. The monitoring process continued for 72 hours and diffusion coefficient was calculated by curve fitting the experimental data and mathematical solution of Fick's law. The obtained diffusion coefficient should be considered as a mean value describing the system of two binders (Karlsson and Isacsson 2002). In next step, maltene phase was extracted, zero-shear viscosity and molecular weight distribution were determined for binders. The results showed that the increase in polarity and molecular size as a result of further distillation reduces the rate of diffusion. They also concluded that the diffusion rate is controlled by viscosity of maltene phase rather than whole binder. Therefore, as aging has small effect on viscosity of maltene, it is postulated that diffusion process is not affected significantly by aging. Moreover the Stocks-Einstein's equation was useful for interpreting the data by FTIR-ATR.

They used a second procedure to determine the diffusion coefficient in the DSR in 2003. Two thin layers of rejuvenator and aged binder were brought to contact using DSR parallel plates and condition then tested. The composite materials were divided to 20 sub-layers to calculate the concentration of rejuvenator in each of the 20 layers. For this purpose they used a mathematical solution of Fick's law and an assumption of a diffusion coefficient. The obtained concentrations were fitted to experimental data using theoretical calculations of viscosity and complex modulus (G^*). Using fitting curves and back calculation of the concentration, a single diffusion coefficient was determined for each temperature. Their findings showed that diffusion determined by FTIR-ATR method happens with change in rheological properties. The results of two methods differ slightly but still are in the same order. They implied that the rate of diffusion in binder is enough to create a homogenous reclaimed binder during the recycling.

Oliver

Oliver (1975) conducted a study on diffusion using tritium labeling of diffusants into asphalt binder. He used two types of oils and dodecylbenzene with three different binders. The goal of study was investigating the possibility of replacing the aged binder at surface of the pavement with oil spray. A 0.5 cm-thick cylinder of asphalt binder was covered by a very thin layer (less than 1 μm) of tritium labeled diffusing material. The samples were sliced after certain times. The radioactivity was inspected in different depths of the binder sample, and diffusion coefficient was calculated. The findings showed that diffusion has an Arrhenius-type relationship with temperature.

$$D(T) = D_0 e^{\left(\frac{k_1}{T}\right) + k_2} \quad [4]$$

He also found that although molecular weight is important for diffusion process, the molecular shape is a greater concern. Moreover data showed the dodecylbenzene with a lower molecular weight diffused faster than the oils with heavier molecular weight (Oliver 1975).

Carpenter and Wolosick

Carpenter and Wolosick (1980) developed a comprehensive study on the rejuvenation process of fresh binder in RAP material. They used extracted binder to examine the rate of diffusion of rejuvenator in the RAP binder. They also performed tests on the storage time of actual mixes to compare the results with binder experiments. In conclusion they described the diffusion process during the rejuvenation to following steps. The high-viscosity film thickness of RAP binder is surrounded by the low viscosity rejuvenator layer. Next, the rejuvenator begins penetrating in the highly-aged binder and as a result of that the mass of covering fresh binder diminishes. After a while, no raw rejuvenating agent remains and the diffusion process still continues. The viscosity of inner layer of the film thickness is lowered due to rejuvenation and at the same time the viscosity of the outer layer is increased. Finally the equilibrium is reached in the film thickness around the aggregate and two binders are fully mixed. However, reducing the viscosity in the interface of aggregate and binder shell may be exempted due to adsorption effects. Moreover, they used staged binder extraction to verify the diffusion process. The staged extraction was performed by first, immersion of asphalt mix in trichloroethylene for 3 min and washing the rest of binder entirely in the second stage. The binder extracted from two stages were recovered and indicated as inner and outer layers of film thickness around the aggregates. From testing the two recovered binders, it was shown that stiffness of outer layer increases while the inner layer softens during the time (Carpenter and Wolosick 1980). Their study also revealed that the

blending between rejuvenator and RAP binder is not limited to mixing time and continues to a more extended time.

Noureldin and Wood

They conducted a similar study with four stages extraction of asphalt binder in 1987. They conditioned the samples for 15 hr at 78°C before the extraction. They immersed samples in trichloroethylene for 5 min in increments from 200 to 700 ml. The results verified Carpenter and Wolosicks' findings (Noureldin and Wood 1987).

2.5 Conclusion from Literature Review

Most of studies on HMA containing RAP demonstrated some extent to high level of contribution of RAP binder in final properties of the mixture. An important concern that may change the performance of HMA containing RAP material is the level of blending between aged and fresh asphalt binders. Both performance and economic competitiveness of HMA produced with RAP material is affected by level of blending. Either accepting or neglecting the blending between fresh and RAP binder will be problematic for final performance of the implemented pavement without any understanding of blending process (Al-Qadi, Elseifi and Carpenter, Reclaimed Asphalt Pavement - A literature review 2007). Either accepting or neglecting the blending between fresh and RAP binder will be problematic for final performance of the implemented pavement without any understanding of the blending process.

A more comprehensive understanding of undergoing interaction of RAP and fresh binder during mixing and implementation is required to assure a more reliable design and construction. However this needs an extensive study on different aspects and factors of using RAP in the HMA to address all concerns. This study will try to demonstrate the interaction between aged

and fresh binder leading to new rheological properties considering effect of time and temperature.

3 Materials and Methods

This chapter will present a detailed description of the materials type, samples preparation, test method, and the procedure used for analyzing experimental data and validating the aforementioned hypothesis.

3.1. Analytical procedure

3.1.1. Materials Preparation

The study aimed to consider a variety of material to verify the hypothesis. The concept of diffusion first was demonstrated using asphalt binders with different chemical structures (i.e., asphaltene contents). The next was applying mortar material from actual and artificial Reclaimed Asphalt Pavement (RAP) material to show the diffusion and blending in practice. The last step was based on testing asphalt mixtures containing RAP, compacted in different temperatures and conditioning times to show the effect of RAP binder on the mixture performance. Based on material used in each step, this section can be divided into three parts:

3.1.2. Asphalt Binder Diffusion Test Sample Preparation

Ideal blending between two materials occurs when both exist in free volumes, are compatible and possess suitable condition to mechanically blend. In practice, a mechanical blending between fresh and RAP binders cannot be expected because the RAP binder in the mix exists as a thin layer covering aggregates obtained from recycling of pavement. However for verifying the diffusion phenomena between two binders using rheological properties, it was necessary to mimic a condition that binders are exposed to each other without any external factor to impose the blending. RAP binder extraction was avoided in this study due to problematic change in

properties (Dan and Bahia 2010). Therefore “Artificial RAP” binder was produced using two or more Pressure Aging Vessel (PAV) cycles of aging. One PAV cycle of asphalt binder is expected to introduce 7-10 years of aging in the field. In this study it is speculated that two or more PAV cycles can provide the binder with enough aging to show similar behavior of actual RAP binder.

Two asphalt binder sources with different chemistries were used in this study, Valero with Performance Grade (PG) of 64-16 and Flint Hills (FH) PG 64-22. The different chemistries for these two binders can be addressed by different asphaltene content where Valero has 9.5% and FH has 16% asphaltene content by weight. Based on previous studies, asphaltene has a significant effect on rheological properties of the asphalt binder. The viscoelastic studies also demonstrated that the diffusion is due to Brownian motion of asphaltene micelles at a sufficient temperature (D. e. Lesueur 1997). Special silicon molds with 25 mm diameter were made in the lab to cast the asphalt samples (Figure 3).

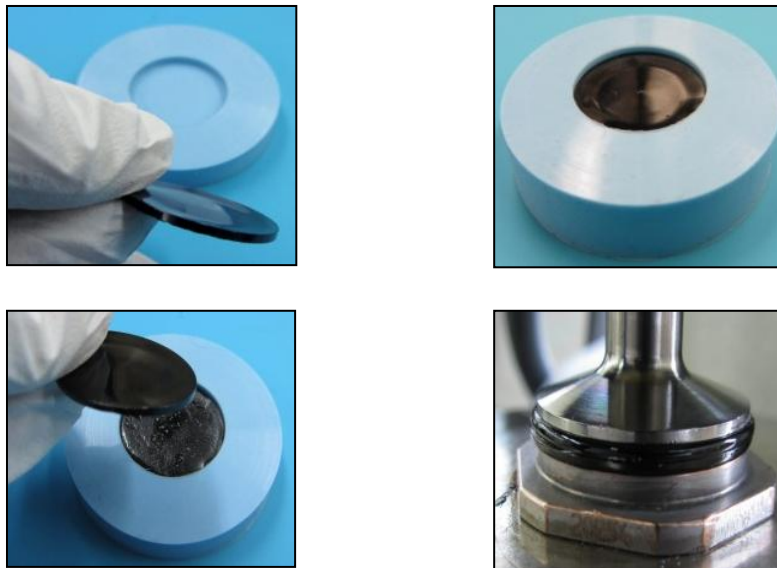


Figure 3 Asphalt binder sandwich, preparation and testing

To control the layers thickness, predetermined amount of hot asphalt binder is poured in the mold and is placed in a flat place to flow with a uniform thickness and cool down. The thickness varies based on the test requirements. Molds are placed in the fridge and after a while the samples are demolded. Asphalt binder sandwiches are made from two asphalt binders with different aging levels. The total required sample thickness in the DSR is 2 mm, however the total sandwich thickness is designed to be slightly more than 2 mm to assure that the required gap size is available.

3.1.3.RAP Mortars

Going one step beyond binder-binder blending, mortars in a smaller scale can demonstrate the effect of blending of RAP and fresh binder in asphalt mixture. Providing and testing of mortars in the lab is more convenient comparing to mixtures. Mortars may not indicate performance characteristics of mixtures, however the blending mechanism can be assumed similar.

The mortars used in this study were provided from two RAP sources and one fresh binder source (Valero PG 64-16). TCP and Stark RAP sources were provided from stockpile of different areas of Wisconsin. Dried RAP was sieved and materials retained on the sieve number 100 (0.15 mm) were collected (Figure 4).



Figure 4 RAP material: (a) Un-sieved RAP material, (b) R #100 RAP, and (c) Burned R #100

To determine the binder content of the fine RAP material, some was burned in the ignition oven based on ASTM D 6307 standard test. The binder content for Stark RAP retained on sieve #100 (R#100) was determined to be 9.2% and for TCP was 6.2%. Mortars with the same total binder content were made of burned R #100 RAP and fresh Valero binder. These mortars were used to eliminate the aging during conditioning in the oven

3.1.4. Artificial RAP Mortar

In a better case scenario for mortars study, it is preferred to have both fresh and RAP binder mechanical properties (i.e. G^*). The RAP binder in the reclaimed asphalt material can be dissolved in chemical solutions such as n-heptane and recovered. This method of separating RAP binder from the aggregate portion is called binder extraction. Performing this study, the binder extraction was avoided due to unknown consequences (Swiertz and Bahia, Tests Method to Quantify the Effect of RAP and RAS on Blended Binder Properties without Binder Extraction 2011). Therefore, “Artificial RAP” was made of aged binder and R #100 granite. The aged binder and granite were heated in the oven and mixed thoroughly. The mortar was continually mixed to cool down, and then placed in a freezer. The mortar pieces were crushed in a mortar and pestle to obtain a granular R #100 Artificial RAP (Figure 5). The total binder content of Artificial RAP was 20% to provide a higher RAP binder replacement in the mortar.



Figure 5 Artificial mortar was provided from R #100 granite and aged F.H binder

To assure that fine aggregates in the mortar are diluted and DSR tests are not confounded by aggregates interaction, some simple test were run to determine the appropriate binder content. Mortars with different percentages of fresh binder in combination with R #100 granite were tested in the DSR (Figure 6).

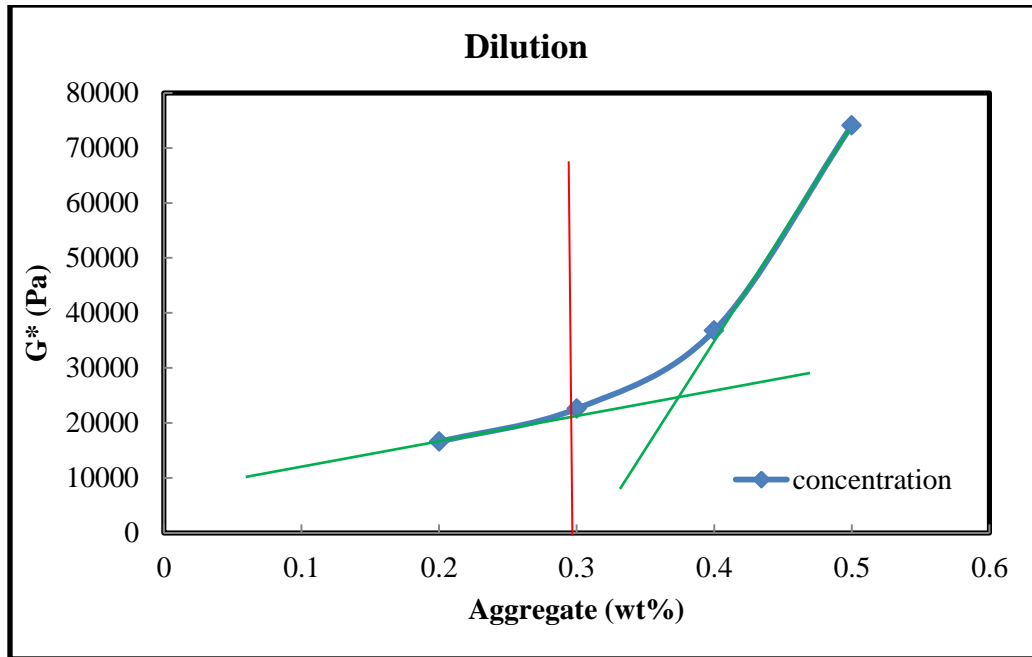


Figure 6 DSR test results for different weight percentages of fine aggregates in the mortar

Based on this test, the region that the G^* starts increasing significantly is the largest concentration of fine aggregate that can be used without any significant interaction between particles.

Determining the maximum applicable fine aggregate content (by weight), fresh F.H binder was added to the Artificial RAP and the same procedure as actual RAP mortars was performed for sampling and DSR testing.

3.2. Test Methods

The different material types, required different testing methods. However for asphalt binders and mortars, the same test methods were used. Asphalt mixtures were tested to evaluate their rutting performance as a comparative base for samples with and without RAP.

3.2.1. Asphalt Binder DSR Diffusion Test

Prepared asphalt binder sandwiches were placed in a calibrated oven to conduct the required conditioning time and temperature. After demolding, the sandwiches are placed in the DSR and a simple rheological test was conducted (Figure 3). For the sake of consistency between mortar and asphalt binder materials, the test specifications were selected the same (Table 2).

Table 2 DSR test specifications

Specification	Asphalt binder sandwich	Mortar
Plate size (mm)	25	25
Gap size (mm)	2	2
Test temperature (°C)	64	64
Strain (%)	2	2
Frequency (rad/sec)	10	10

The strain was selected low because of the stiff nature of mortars which requires low strain levels in DSR testing. The frequency of 10 rad/sec is normally used for measuring G^* in the DSR.

3.2.2. Mortar Samples

As demonstrated in Table 2 the same specifications were used for DSR test on mortars. It is necessary to avoid the fine aggregates interlock during oscillatory test in the DSR. For this purpose, a minimum gap size is required. The requirement for a proper gap size is defined as a

thickness of larger than one fourth ($>1/4$) of largest aggregate size (AASHTO T 315). The mortar contains the maximum aggregate size of 0.30 mm (sieve #50) which indicates the satisfactory gap size of 2mm.

3.3. Theoretical Stiffness Calculations

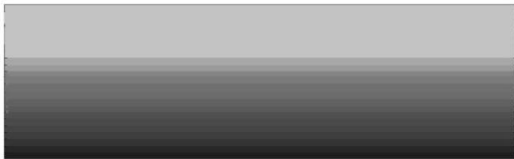
The analytical procedure used in this study is according to the theory of composite materials. Based on changes in composite structure, the mechanical properties vary. Complex shear modulus (G^*) is a good indication of binder mechanical properties which has a widespread use in asphalt binder gradation. Asphalt binder sandwiches described in previous sections, in initial case are considered as a two-phase composite, fresh and aged binder. The thickness of two binders is assumed to be equal and a proper bond is provided by DSR pressure for unconditioned samples. Based on the theory of diffusion, two binder phases will start blending in the intermediated boundaries if suitable conditions are provided. In this case a third phase which is a mix of fresh and aged binder will form in the composite. As the diffusion is time and temperature dependent, the diffusing phase will gradually diffuse in the medium and the gradient of the diffusing substance is not uniform in the entire material thickness. Therefore the procedure assumes the composite is formed of a layer of fresh binder (as diffusing material) and an aged binder medium with gradient of fresh binders' concentration in it which changes with time and temperature. In other words, during the conditioning the fresh binder thickness will continuously decrease while the blended binder thickness will increase until equilibrium between two binders is achieved. By dividing the aged binder layer to sub-layers an accurate enough analysis is achievable (blended binder layers with different concentrations of fresh binder will be considered in the analysis). The equilibrium state is an indication of full blending between two binders forced by diffusion phenomena.



a) *Fresh and aged binder without blending*



b) *Blended binder regions start forming due to diffusion of fresh binder in the aged binder layer.*



c) *Along with blending progress the concentration of fresh binder in blended layers increases*



d) *Aged binder layer is dissolved in the blended binder layer*



e) *An equilibrium is reached between two binders (full blending)*

Figure 7 Schematic of change in concentration of fresh binder in aged binder due to diffusion phenomena

By dividing the aged binder into layers, the theoretical stiffness of the composite can be calculated as following:

$$\frac{h_T}{G_T^*} = \frac{h_{fresh}}{G_{fresh}^*} + \frac{h_1}{G_1^*} + \frac{h_2}{G_2^*} + \frac{h_3}{G_3^*} + \dots \quad [1]$$

Where:

G_T^* : The total stiffness of the composite

G_{fresh}^* : The stiffness of the fresh binder

$G_{1,2,3,\dots}^*$: The stiffness of sub-layers in the aged binder

h_T : The total thickness of the sandwich

h_{fresh} : The thickness of the fresh binder

$h_{1,2,3,\dots}$: The thickness of sub-layers

Considering the mentioned hypothesis for diffusion theory, for a sandwich of fresh and aged binder, h_{fresh} decreases while $h_{1,2,3,\dots}$ increases during conditioning at a high enough temperature.

Although the initial stiffness of aged binder was determined using a simple test in the DSR, for sub-layers it was required to estimate the stiffness changes while the fresh binder was diffusing in. For a given concentration of two binders, the viscosity can be calculated using Equation 2 a mixing rule which was first presented by Arrhenius (1887) and then validated for asphalt binders by Davison et al. (1994).

$$\eta_{mix} = \frac{\eta_A^\alpha \cdot \eta_B}{\eta_B^\alpha} \quad [2]$$

Where, η is viscosity of asphalt binders, A and B are indications for two asphalt binders and α is the concentration of the binder A. As there is a direct relationship between viscosity and stiffness changes, for G^* of two binders the Equation 2 can be rewritten as following:

$$G_{mix}^* = \frac{G_A^{*\alpha} \cdot G_B^*}{G_B^{*\alpha}} \quad [3]$$

However, in this project a test was conducted to show the correlate the G^* changes with concentration of one binder in another. Aged and fresh binders were heated to 140°C separately, and then different amounts of aged binder were added to the fresh binder in a silicon container. Using the silicon container is critical to assure that binders do not stick to the container and designed concentration is accurate enough. The combinations are mixed manually in two or more cooling and heating cycles to produce uniform mixes so called *Mechanically Blended* in this project.

Samples from *Mechanically Blended* mixes were tested in DSR and the G^* changes versus aged binder concentration were plotted (Figure 8). The trend of G^* changes shows a very good correlation with an exponential equation.

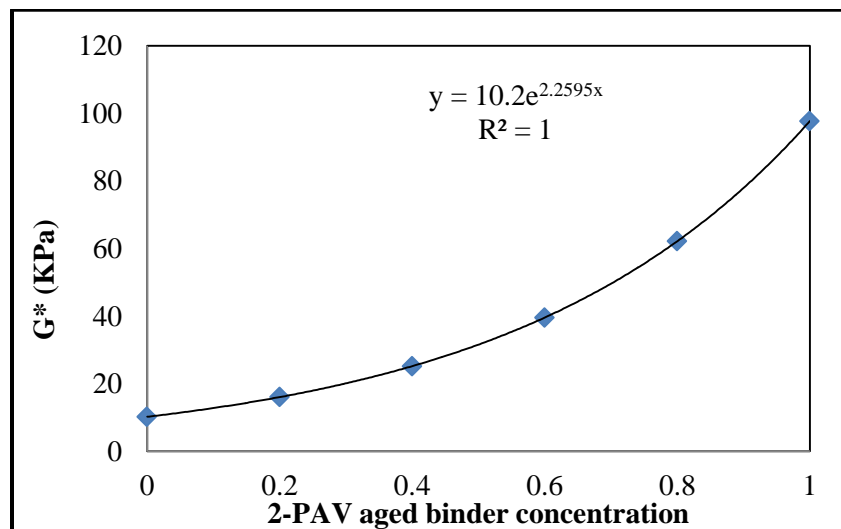


Figure 8 Trend of G^* changes for different concentrations of aged binder in the mix

Results also were presented in logarithmic scale which showed a linear correlation between logarithm of the G^* and concentration of aged binder (Figure 9). This test was repeated for two

binder sources and the linear correlation between Log G^* and concentration was observed for both.

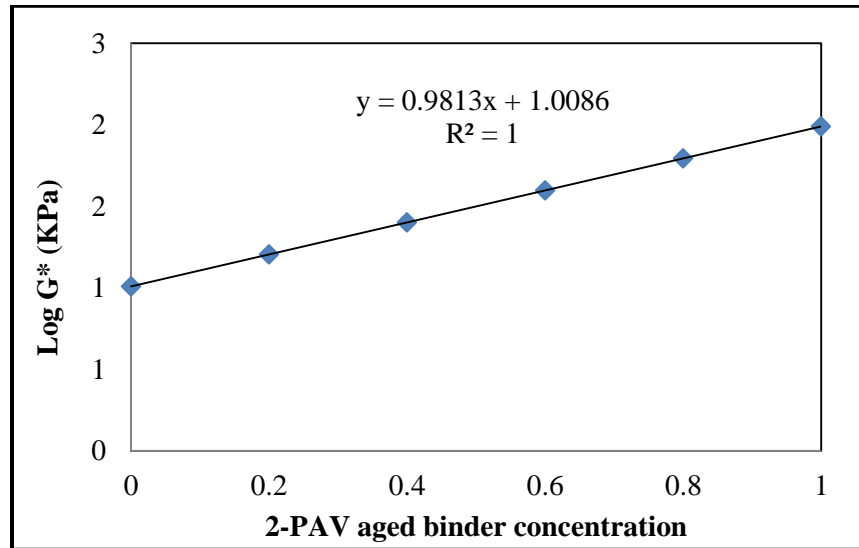


Figure 9 Linear correlation between logarithm of G^* and concentration

Findings from this test shows that the G^* of a two-binder-mix with any concentration can be obtained by having the stiffness of both binders. As it can be determined from Figure 9 for any concentration of aged binder, the G^* of the mix can found.

3.4. Proposed Analytical Procedure for Diffusion Calculations

As the fresh binder diffuses into the aged binder, the concentration of aged binder in each sub-layer changes. According to this assumption, the thickness of the sub-layers changes as well. Using the experimental findings or theoretical equation for *Mechanically Blended* mixes, the stiffness of each sub-layer can be estimated based on the diffusion progress. By placing the values obtained for sub-layers in the Equation 1, the total theoretical G^* for the composite is determined.

Knowing the theoretical stiffness of the composite and effect of concentration change, the diffusion rate should be considered as the only unknown in the procedure. For this purpose a solution of the Fick's law developed by Crank was considered based on the sample geometry and boundary condition (equation 4) (Crank 1975). Based on this equation the initial concentration of fresh binder C_0 in the Artificial RAP binder wafer is zero and is one in the fresh binder wafer. It is also assumed that there is no mass loss, the binder does not diffuse into the plate, and the geometry of the sample does not change. The Aged binder layer can be discretized into n sub-layers and the concentration of fresh binder $C(x,t)$ in each Artificial RAP binder sub-layer can be calculated in conditioning time steps t (similarly, the concentration of Artificial RAP binder could be calculated in sub-layers of the fresh binder wafer). Assuming a constant diffusion coefficient D , an analytical solution to Fick's Second Law can be written to satisfy the boundary conditions of this problem (Equation 2) (Crank 1975).

$$C(x, t) = \frac{4C_0}{\pi} \sum_{n=0}^{\infty} \frac{1}{2n+1} e^{\left(-\frac{D(2n+1)^2 \pi^2 t}{l^2}\right)} \sin \frac{(2n+1)x\pi}{l} \quad (4)$$

Where:

C_0 : Initial concentration of fresh binder, [%]

n : Sub-layer, [1, 2, 3, ..., 10]

t : conditioning time, [sec]

l : 'Artificial RAP' binder layer thickness, [1 mm]

x : Vertical distance from the bottom DSR plate to the top of each sub-layer n , [mm]

D : Diffusion coefficient, [mm^2/sec]

Although the diffusion coefficient is assumed constant for this work, Equation 1 suggests that the diffusion coefficient not only changes with each specific molecule that diffuses, but also with

time as the medium viscosity and concentrations change. However, treating it as constant provides a suitable average as we are measuring a bulk material property that is not associated with any one molecule. The concentration of fresh binder within the Artificial RAP binder at any conditioning time is then obtained from the summation of concentration in all sub-layers and subtracting from the overall fractional concentration of diffusing binder in entire wafer thickness (i.e., since the fraction of fresh binder is equal to the Artificial RAP binder, the fractional concentration is $0.5C_0$) (Figure 10).

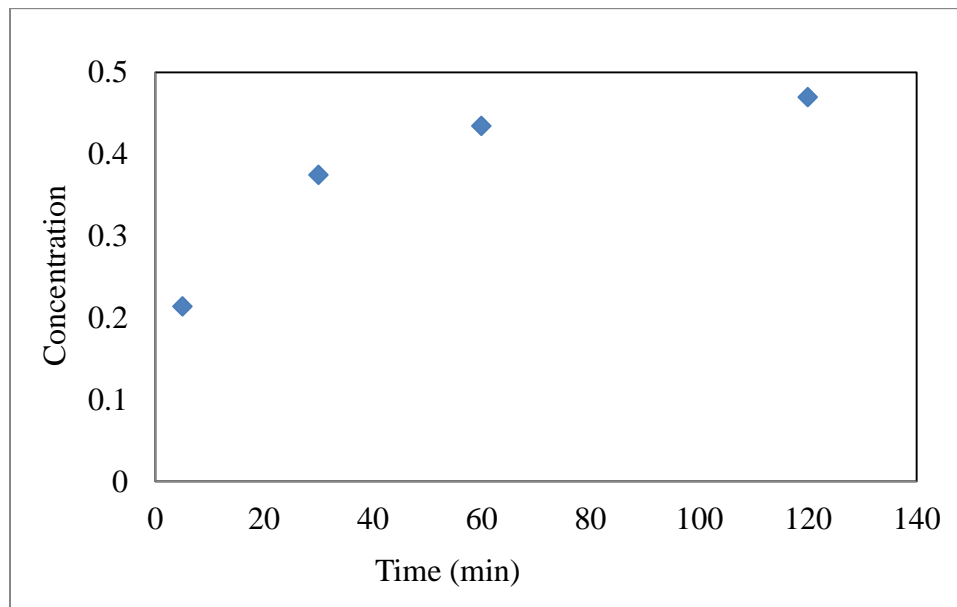


Figure 10 Example of concentration change with time based on Fick's law equation for a defined diffusion coefficient

As demonstrated in Figure 10 using a defined diffusion coefficient the concentrations versus time can be obtained from Fick's law. The concentration can be calculated for each sub-layer, then based on results from Mechanically Blended samples, and Equation 1, a relation between G^* and conditioning time can be established (Figure 11). The composite theory used for the binder sandwich demonstrates that the more the fresh binder diffuses into the aged binder, the

higher the stiffness of the composite gets. Therefore it is expected that a similar trend as concentration-time can be observed for G^* -concentration.

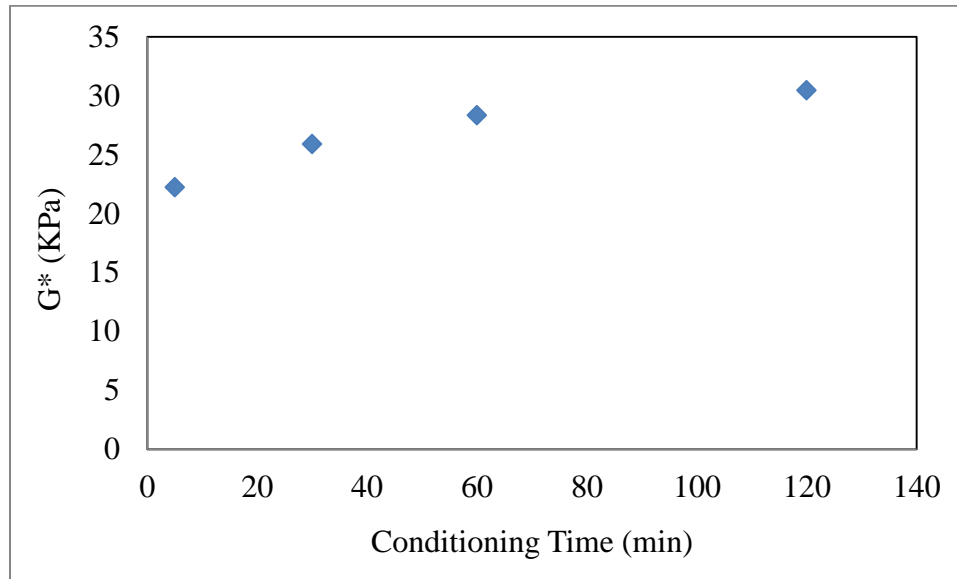


Figure 11 Stiffness change vs. conditioning time, obtained from Fick's law and composite theory calculations for F.H binder at 120 C

Figure 11 shows typical results from Fick's law calculations correlated with the Mechanically Blended samples results and the applied composite theory. A spreadsheet was developed for calculations of concentration during time based on Fick's law and a multilayered analysis

Experimental data for sandwich samples, are presented in terms of G^* and Conditioning time. Each sandwich is conditioned at a specific temperature for a determined time and prepared for testing in the DSR. Once the results are plotted as G^* versus conditioning time, experimental data can be matched with plot obtained in previous step if a proper diffusion coefficient is selected.

The spreadsheet uses an iterative procedure to minimize the sum of squares of difference between two G^* values (theoretical value from Fick's law and experimental output) to estimate the proper diffusion coefficient. The analysis procedure for diffusion calculations is summarized in Figure 12.

3.5. Desired time and temperature to acquire proper blending

The other objective of this research was to determine if a full blending between fresh and RAP binder can be expected. The desired conditioning time and temperature to acquire an acceptable blending level in mixtures containing RAP material will be time and cost effective.

This stage of work was mostly based on mortar testing, however asphalt binder sandwich method was applied to answer some questions as well.

3.5.1. Analyzing mortar testing results

In the initial work plan, two Rap sources were evaluated. It was hypothesized that the RAP source can show different blending behavior (trend of stiffness changes) due to different aging level of the containing RAP binder. The samples were prepared according to the procedure described in Materials section. Fresh binder was added to the R #100 RAP aggregates and conditioned for different times in three specified temperatures (100, 120, and 140°C). The same aggregate sources were burned in the ignition oven and mortars with the same binder content were provided. The mortars without RAP binder were used to inspect the effect of aging during the conditioning in the oven. The aging effect should be eliminated from stiffness changes of the unburned samples to make sure that the results are not confounding by the binder aging during the conditioning.

The changes in stiffness for different conditioning durations can be a demonstration of blending progress in the mortar. The trend of changes also can be used to derive a conclusion about the proper blending time and temperature.

3.5.2. Evaluating the film thickness effect

Another RAP source specification which was speculated to affect the blending time, was the film thickness of both RAP and fresh binders. The film thickness is the binder content divided by the total surface area of aggregates and the filler. In this study the film thickness effect was investigated using different thicknesses of fresh and aged binders in the binder sandwich method (Table 3).

Table 3 Film thickness test using asphalt binder sandwiches

Asphalt binders		Thickness- mm		Conditioning	
Fresh	2-PAV	Fresh	2-PAV	Temperature ° C	Time (min)
FH	FH	1.5	0.5	145	1, 5, 30, 60, 120
		1	1	145	
		0.5	1.5	145	

4 Results and Discussion

4.1. Binder Blending

Evaluating the effect of time and temperature on blending between fresh and aged binders was speculated to reveal useful information to address the blending in asphalt mixtures containing RAP. Although the thickness of asphalt binder sandwiches was much larger than film thickness of RAP and fresh binders in the mixture, the time and temperature dependency of the blending progress was of great interest.

4.1.1. Binder Sandwich

Conditioned asphalt binder sandwiches were tested based on the specifications introduced in previous section. The results were plotted in terms of G^* and conditioning time for different temperatures. In the initial work plan, it was tend to provide sandwiches from two different binder sources. Fresh Valero was exposed to 2-PAV F.H and fresh F.H was conditioned with 2-PAV Valero.

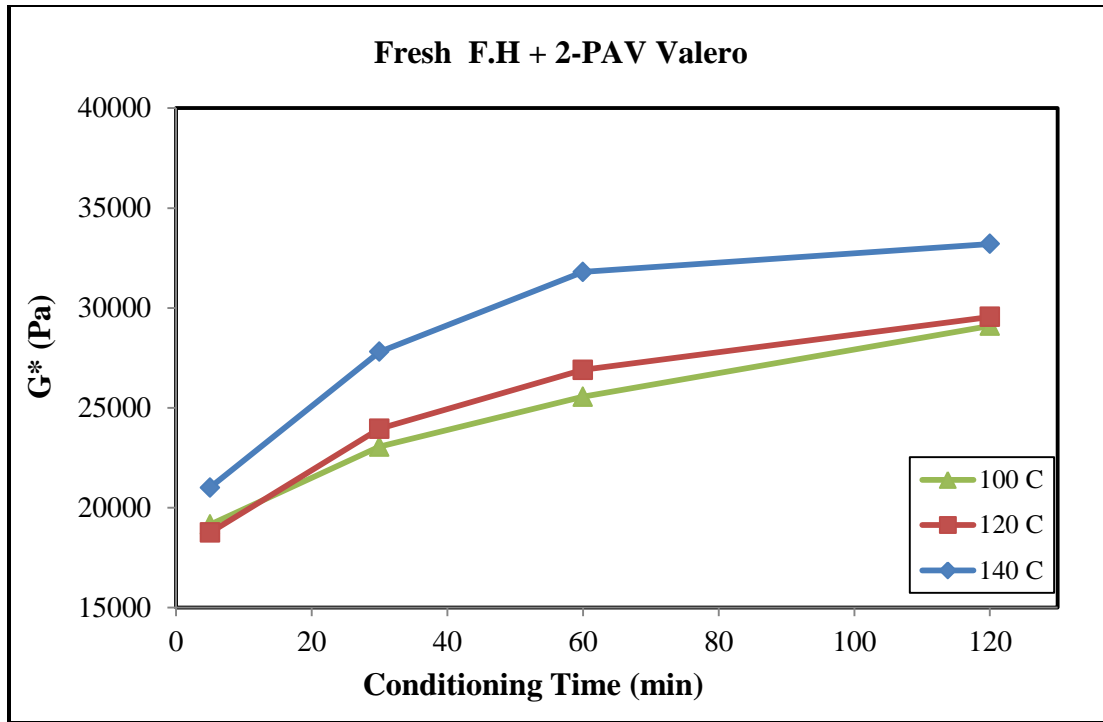


Figure 13 Fresh F.H and 2-PAV aged Valero sandwich at different temperatures

Figure 13 shows the DSR results for fresh F.H and aged Valero sandwich at three temperatures. The trend demonstrates increase in stiffness. Based on the composite theory explained in the previous section, as the thickness of fresh binder in the sandwich decreases, the total measured G^* increases.

The trend demonstrates a higher rate of stiffness changes for conditioning at 140°C while at lower temperatures (120 and 100°C) the changes are lower and almost the same as each other. It seems that the effect of fresh binder G^* (or viscosity) on blending is less significant for moderately lower temperatures (in this case 100 and 120°C). Literature recommends a noticeable effect of the binder viscosity on the diffusion phenomena and as result the blending between two binders. The viscosity results for two binders will be presented later.

The other notable demonstration of blending trend is the higher rate of blending (G^* increase) at initial times. Considering G^* of 18,470 Pa for a sandwich without conditioning time, about 65% of stiffness (blending) of two hours conditioning occurs in first 30 minutes for samples conditioned at 140°C. This percentage for 100°C and 120°C in the same time range is 43 and 54 respectively. These observations can be explained by the different molecular mobility of asphalt binder in different viscosities. According to Rotational Viscometer (RV) measurements, there is significant difference between fresh binders (diffusing material) viscosities at different temperatures (Table 4). According to the literature, the Brownian motion of asphalt binder molecules is facilitated at lower viscosities (D. Lesueur, The colloidal structure of bitumen: Consequences on the rheology and on the mechanisms of bitumen modification 2009).

Table 4 Fresh binder dynamic viscosities at different temperature

Temperature (°C)	Viscosity (Pa.s)	
	Valero	F.H
100	3.72	3.92
120	0.93	1.00
140	0.32	0.35

Based on the theory explained in chapter 3, the stiffness of fully blended mix of two materials can be estimated using either the mechanically blended testing method or the modified Davison mixing rule (Equation 3). Applying these methods, the G^* of a fully blend of equal portions of fresh F.H with 2-PAV Valero is 32,000 Pa. The test results for samples conditioned at 140°C shows a full blending after two hours (Figure 14). However the full blending cannot be achieved for lower temperatures even after two hours of conditioning.

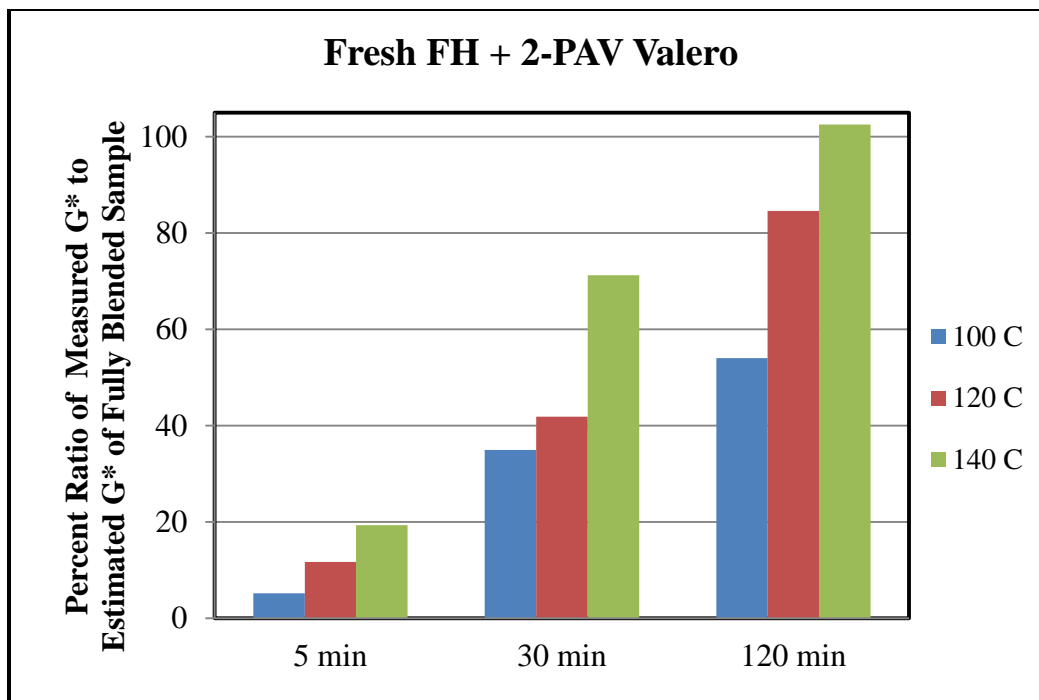


Figure 14 G^* measured for samples at different conditioning time and temperatures as percentile of estimated G^* of a fully blended sample

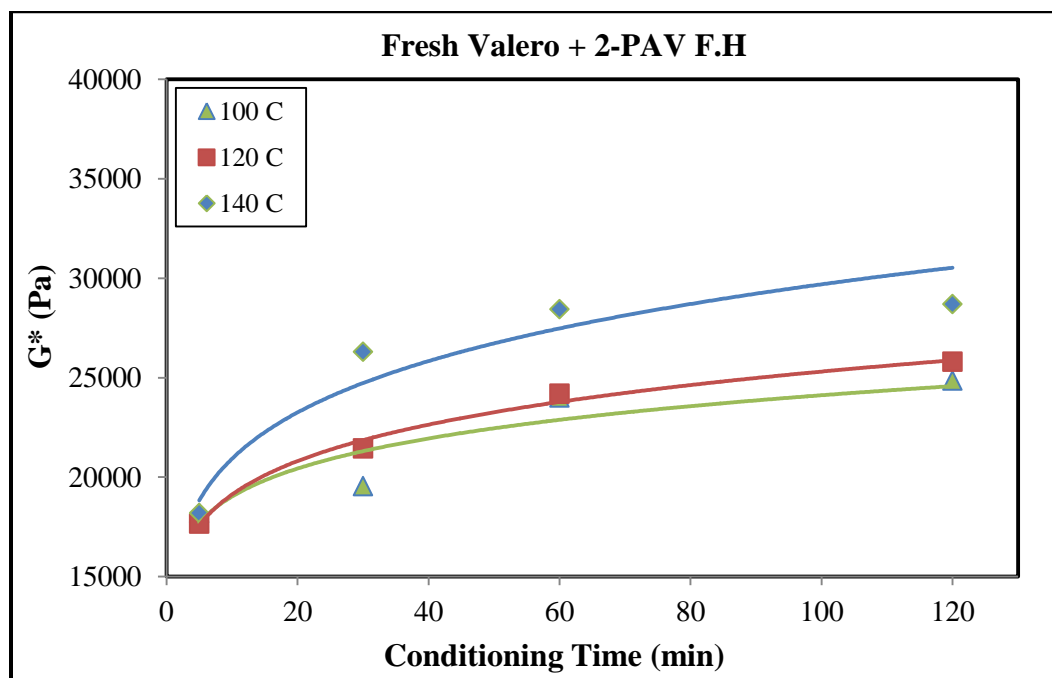


Figure 15 Fresh Valero and 2-PAV aged F.H sandwich at different temperatures

A similar trend was observed for binder sandwiches made with fresh Valero and aged F.H. however the rate of blending seems to be lower for this combination. For two lower temperatures again a very slight difference was noticed.

Analyzing the same as fresh F.H and aged Valero, fresh Valero in combination with aged F.H shows slightly different behavior. The blending (or G^* increase) in first 30 minutes of conditioning comparing to the total two hours conditioning is 29%, 58% and 79% for conditioning temperatures 100°C, 120°C, and 140°C respectively. Comparing to the estimated blending G^* (using Equation 3.3) of 30,200 Pa, a nearly full blending also was observed after two hours conditioning at 140°C (Figure 16).

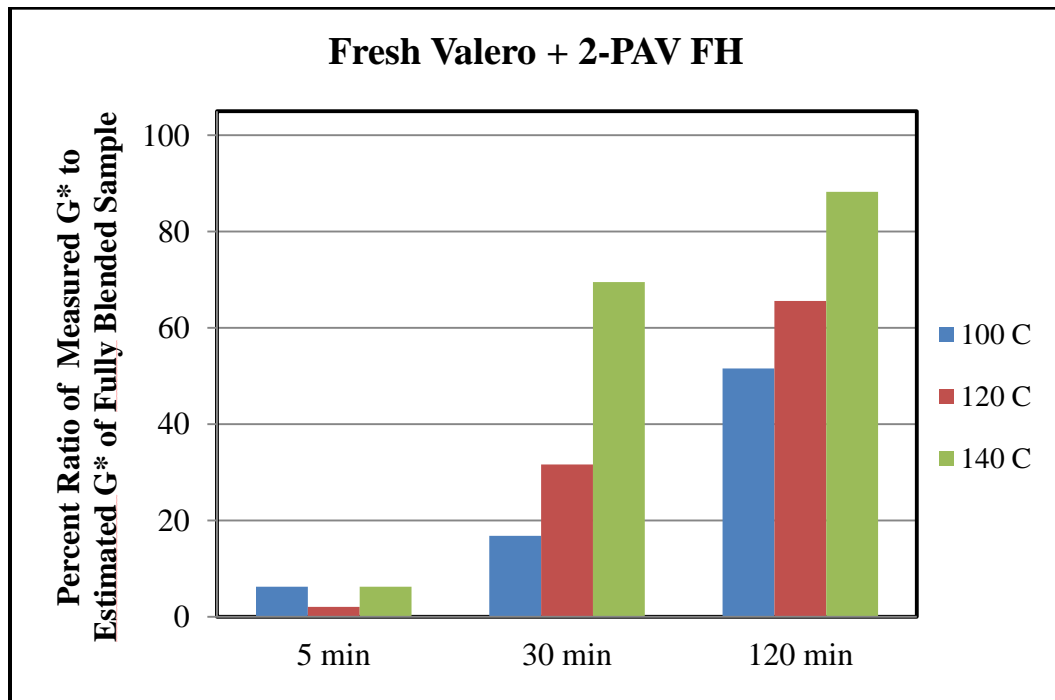


Figure 16 G^* measured for samples at different conditioning time and temperatures as percentile of estimated G^* of a fully blended sample

The increasing trend of G^* at 140°C is milder comparing to the fresh F.H and aged Valero. This might be explained by the different chemical composition of two fresh binders as which are assumed to be the diffusing materials.

4.1.2. Mortar

Based on the hypothesis and work plan it was necessary to investigate the blending between fresh and RAP binders in practice. To monitor the effect of RAP binder on the final properties of the binder in the mixture, it was more favorable to present the blending behavior in terms of G^* . For this purpose sieved RAP material were mixed with fresh binders and conditioned in the oven. The same RAP material was burned and obtained aggregate particles are mixed with binder based on the same total binder content. Mortars without RAP were used to eliminate the effect of the aging during the conditioning in the oven and estimate the initial effect of RAP binder on the mortar stiffness. All presented results are corrected for aging effect.

The first RAP source used in this study was a stockpile from Stark Company in Milwaukee. The total binder content of R #100 RAP material was determined to be 9.2%. Figure 17 shows the typical DSR results at three different temperatures. A higher blending is observed for samples conditioned at 140°C . The trend shows a high possibility of full blending after two hours of conditioning. On the other hand the samples conditioned at lower temperatures exhibit a lower blending rate. It seems at lower temperatures, the total RAP binder is not available to provide a higher blending level.

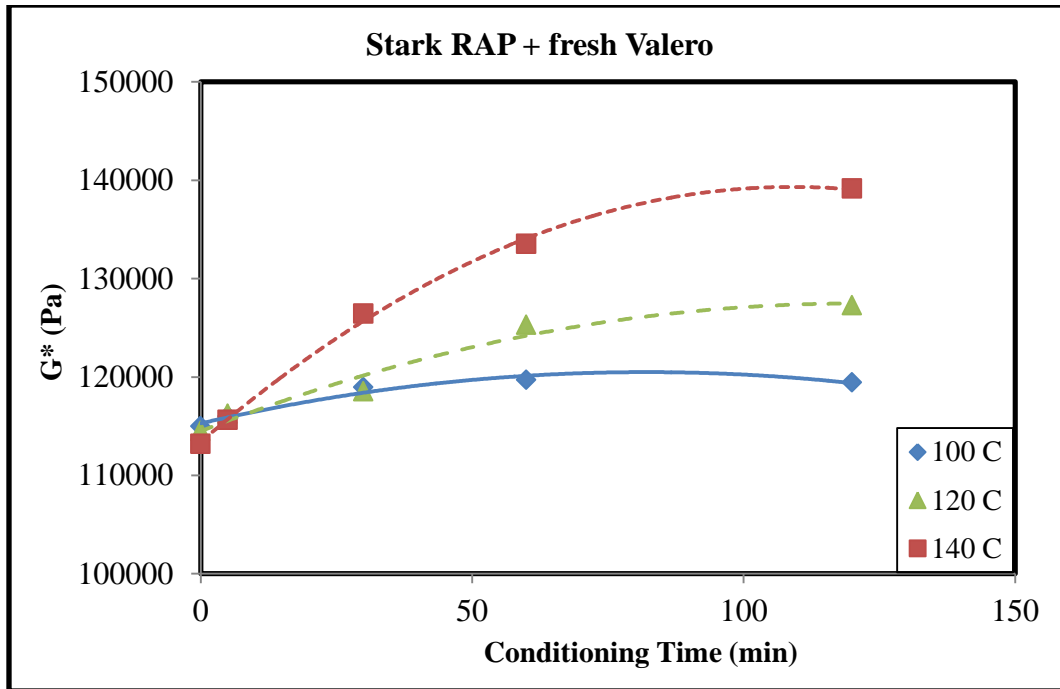


Figure 17 DSR results for Stark RAP mixed with fresh Valero conditioned at different temperatures

Comparing the G^* for samples instantly taken from mixes with and without RAP mortar, there is a significant effect of RAP binder on the stiffness even at very short time after mixing. The stiffness of mortar sample with RAP exhibited a 1.4 times larger G^* than the sample with no RAP binder (burned RAP mortar). This effect can be related to the instant stiffening effect of RAP binder on the fresh binder and the composite layers formed around the fine aggregates. The test also demonstrated that the effect of conditioning after two hours increases the total G^* as much as 20% of the G^* of initial samples (samples without conditioning) (Figure 18). This may indicate that most of the blending happens instantly after mixing and effect of conditioning for further times can be negligible.

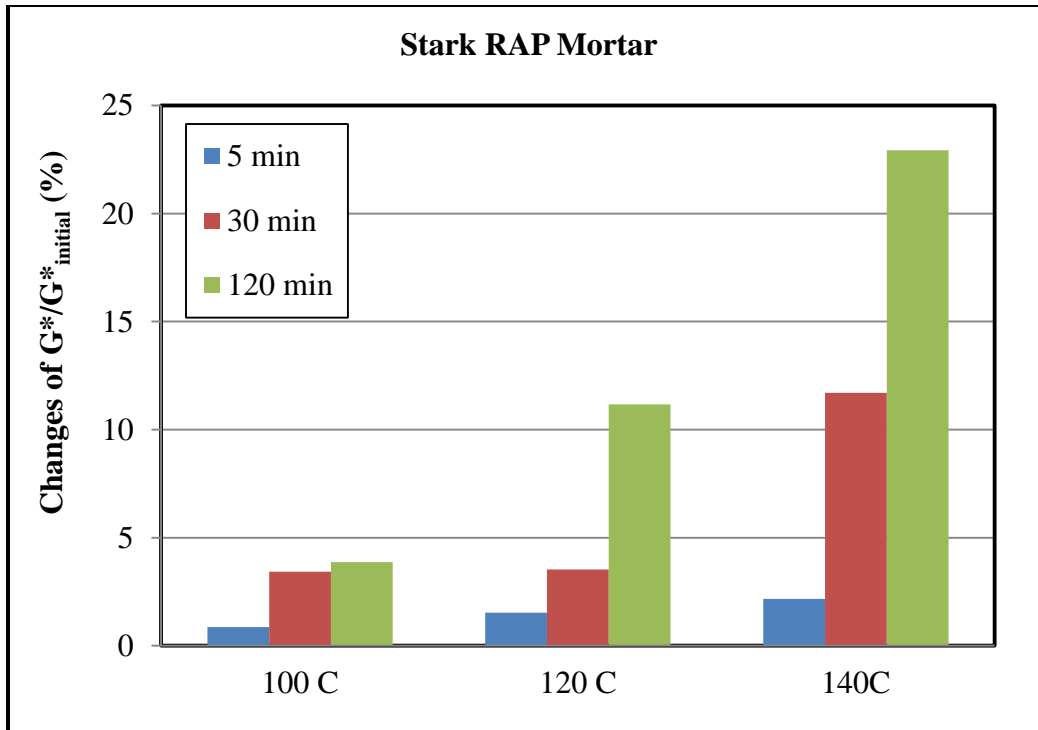


Figure 18 Changes of G^* at different conditioning time and temperatures comparing to samples with no conditioning (Stark RAP Mortar)

The TCP as another Rap source was used in combination with fresh Valero binder. The binder content of material retained on sieve #100 was 6.2% which was lower than binder content of Stark RAP source. The results show similar G^* changes for samples conditioned at 120°C and 140°C. The trend demonstrates small stiffness changes for samples conditioned for 60 and 120 minutes. The blending trend for samples conditioned at 100°C is very close to a flat line which indicates the temperature sensitivity of the blending for this source.

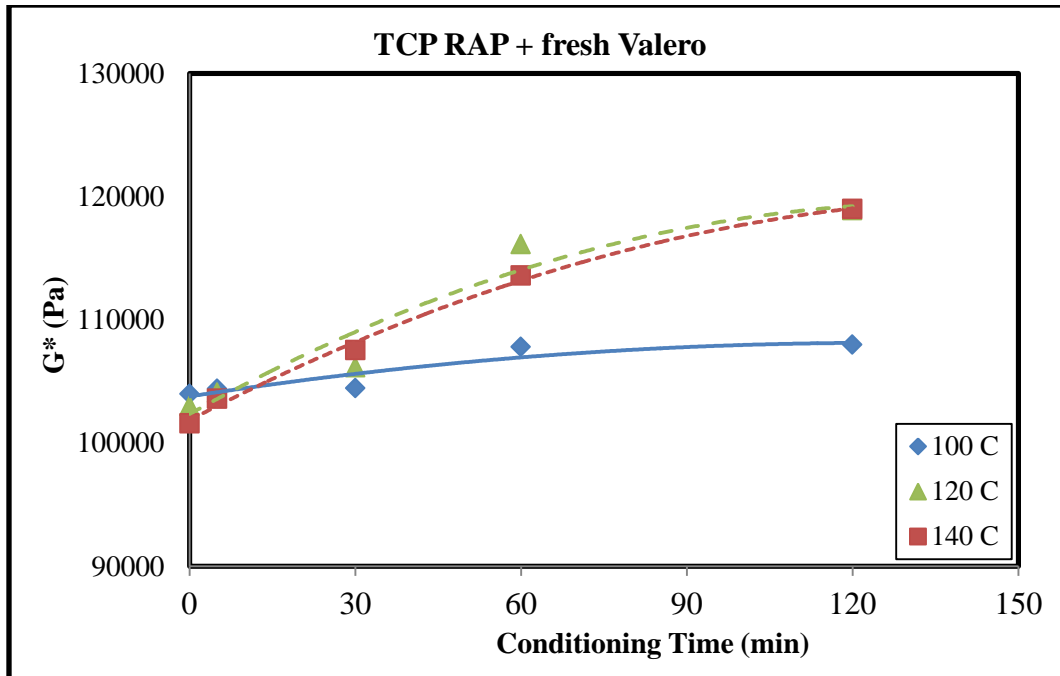


Figure 19 DSR results for TCP RAP mixed with fresh Valero and conditioned at different temperatures

Performing the same analysis for TCP RAP mortar, the existence of the RAP binder in the mortar, causes an instant increase by 1.37 times of the mortar made with the burned RAP aggregates. Conditioning time for two hours increased the G^* just by 17% (Figure 20).

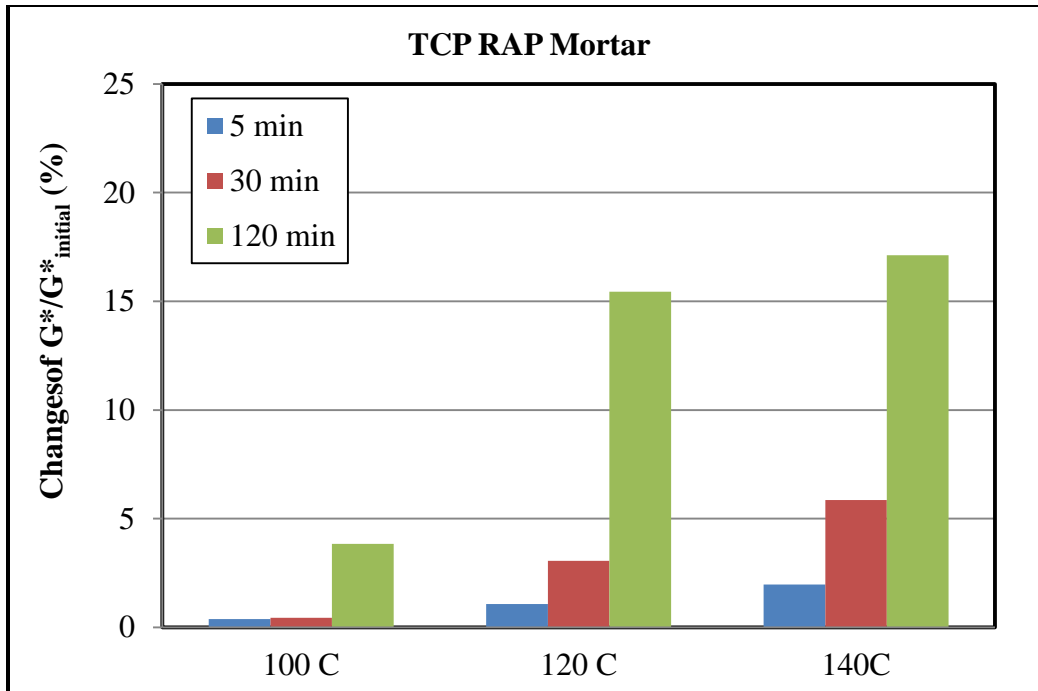


Figure 20 Changes of G^* at different conditioning time and temperatures comparing to samples with no conditioning (TCP RAP Mortar)

Comparing two RAP sources indicates that different blending behavior is expected for various stockpiles even using a unique fresh binder source.

Regardless of the promising repeatability of mortar samples, there were still doubts about the blending determined using the DSR results. The binder adsorption was speculated to be a problem confounding the results. The possibility of added fine particles (such as ash or binder residue) after burning RAP aggregates in the oven which could lead to a changed size distribution was also a concern. The size changes could be inevitable due to degradation of aggregates in very high temperature of ignition oven (550°C). To imitate this concern and provide a full control on all materials specifications, artificial RAP was produced in the lab.

The artificial RAP was mixed with fresh F.H and conditioned at two temperatures (100°C and 145°C). Samples were taken in the specified time domains and instantly placed in the freezer to avoid more blending and especially settlement of fine aggregates in the binder.

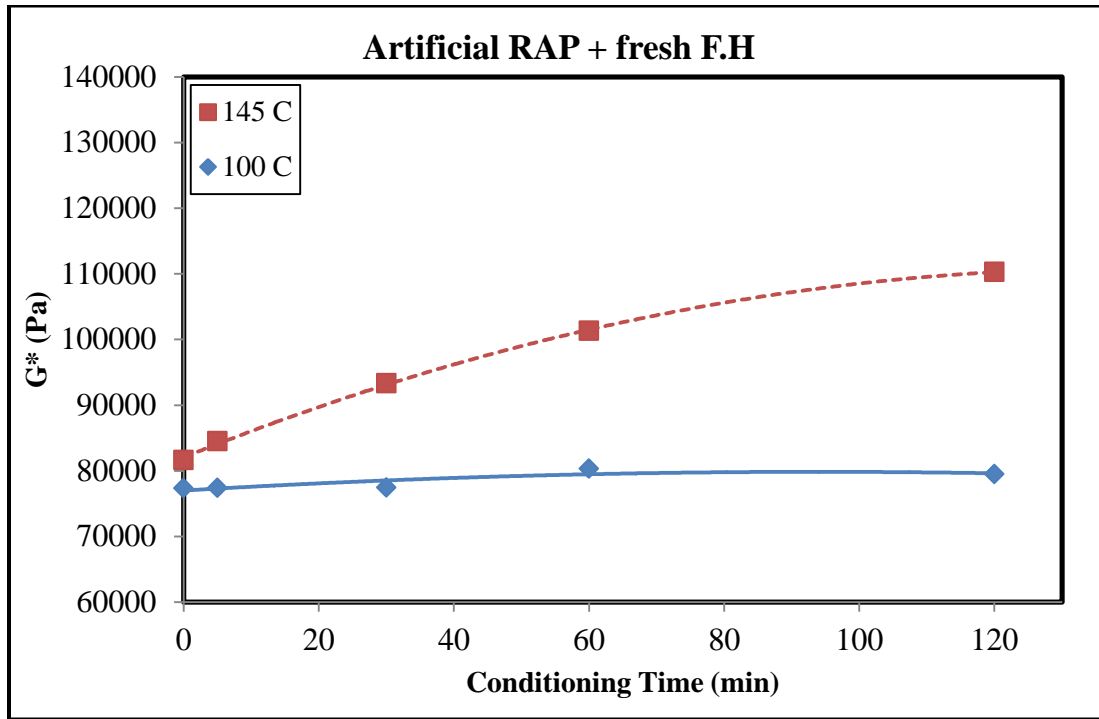


Figure 21 DSR results for Artificial RAP mixed with fresh F.H at 100 C and 145 C

Test results were comparable with trends observed for actual RAP material although the binder contents are very different. Samples conditioned at 100°C show very slight G^* increase for higher conditioning durations. The effect of the RAP binder in the mortar caused 282% G^* increase compared to the granite R #100 with the total fresh binder content of 48%. The effect of conditioning for artificial RAP was determined to be more significant comparing to the actual RAP material. This might be due to the higher RAP binder content of the artificial RAP mortar which requires more time to provide a full blending.

Based on the study observations, it can be speculated that the effect of temperature on blending in mortars containing RAP is decreasing the viscosity of fresh binder and softening the RAP binder to provide suitable condition for blending between two binders. Therefore at higher conditioning temperatures, higher rates of blending and higher maximum achievable blending is plausible.

4.2. Diffusion calculations

Following the analytical procedure outlined in chapter 3, the fitted diffusion coefficient D was calculated. Asphalt binder sandwiches made of different fresh and aged binders were used in the procedure. Based on the testing conditions, the diffusion coefficient was calculated for binder sandwiches at three conditioning temperatures. Some assumptions were made to calculate the diffusion coefficient for mortars as well.

4.2.1. Asphalt Binder

A spreadsheet was developed to calculate the diffusion coefficient. To fit the Fick's law G^* -time results to experimental data and calculate the diffusion coefficient, first the G^* of fresh and aged binders should be inserted to the spreadsheet. Then the DSR sandwich test results for asphalt binder sandwiches should be entered to the specified column. Finally the D (mm^2/sec) will be calculated using minimizing the sum of squares.

Figure 22 shows the Fick's law results fitted to the experimental data using the spread sheet for fresh FH and 2-PAV Valero sandwiches at three different temperatures.

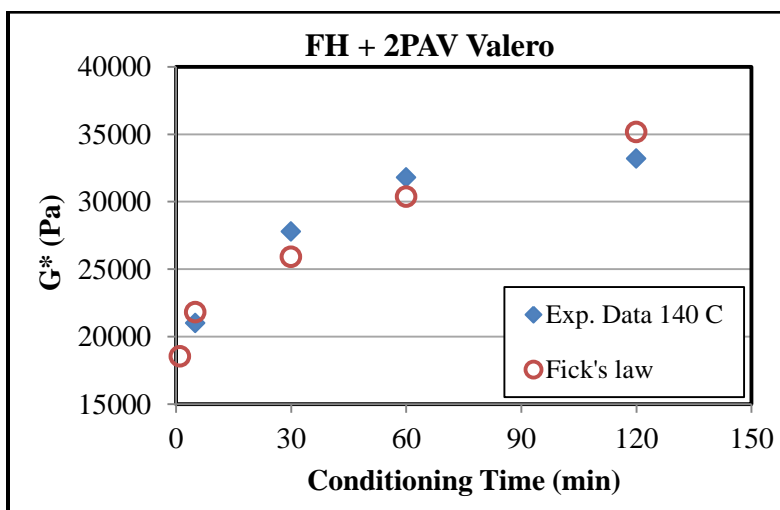
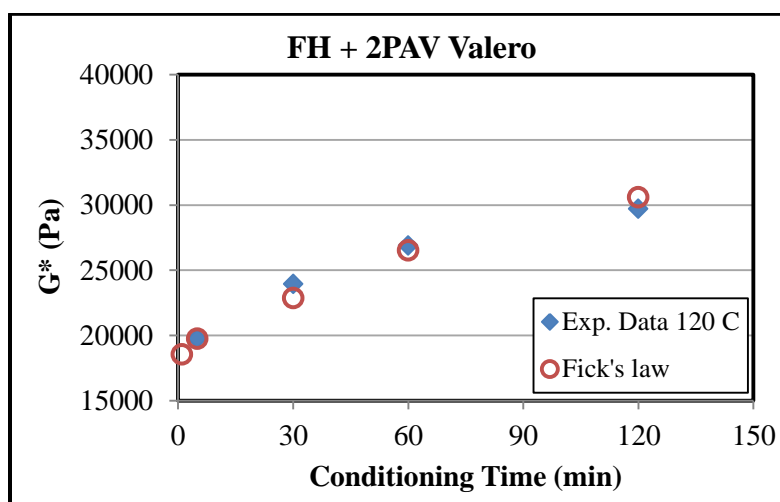
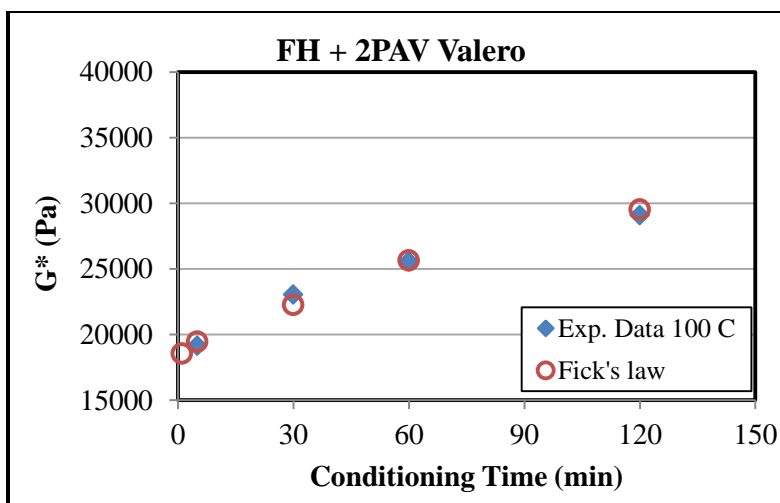


Figure 22 Fitted Fick's law calculations to DSR measurements

As it is demonstrated, a good correlation was made between DSR measurements and Fick's law calculations. Layered system for aged binder (10 sub-layers instead of one) used to estimate the composite G^* which fitted very well with experimental results. The fresh binder concentration changes due to diffusion, also was estimated in 10 sub-layers of the aged binder layer. Therefore, the appropriate stiffness was calculated for each sub-layer based on the diffusion coefficient and the conditioning time.

With the same considerations, diffusion coefficients were estimated for fresh Valero with 2-PAV FH sandwiches as well. The results were in the orders of 10^{-7} and 10^{-6} mm²/sec. Comparing the diffusion coefficients for two sets of binder sandwiches (Figure 23), fresh FH shows slightly higher diffusion rate in 2-PAV Valero comparing with to the other combination.

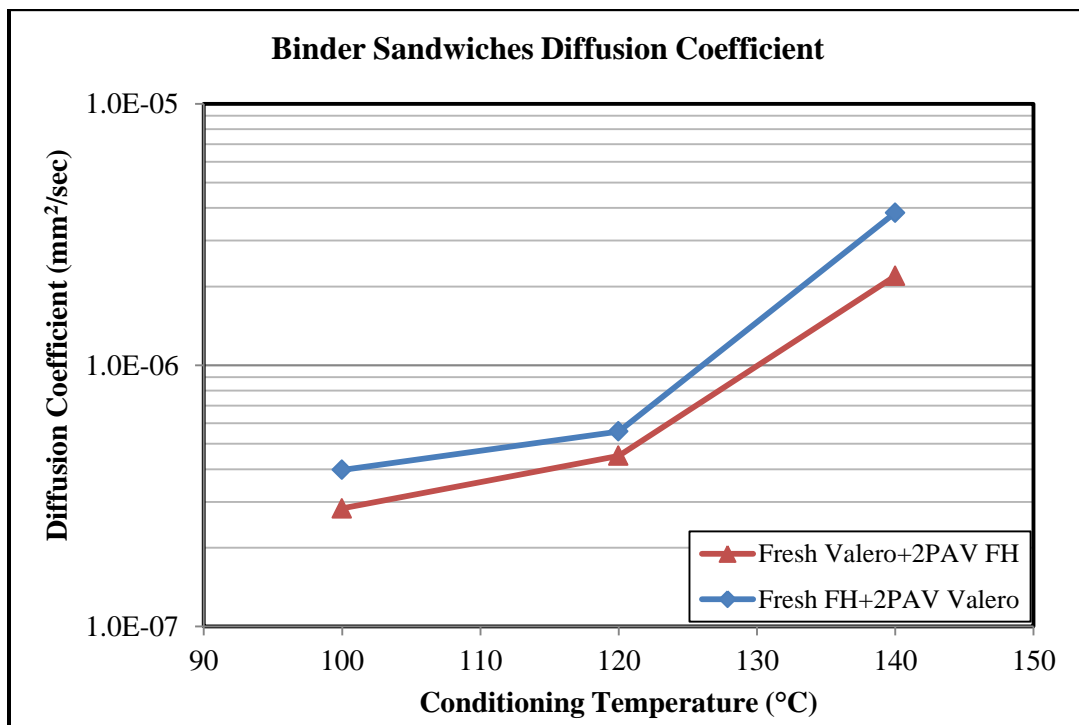


Figure 23 Diffusion results for two binder sandwich combinations

Based on literature (D. Lesueur, The colloidal structure of bitumen: Consequences on the rheology and on the mechanisms of bitumen modification 2009), it was expected to observe slightly higher diffusion rates for fresh Valero comparing to fresh F.H binder due to lower dynamic viscosities at different temperatures (Table 4). Therefore, regarding to different chemistries (different asphaltene contents), it can be speculated that factors other than just viscosity contribute to diffusion phenomena.

However, by comparing different temperatures results the diffusion coefficients were found to be highly dependent on the viscosity. For both combinations, the diffusion rates are found to be close at 100°C and 120°C while a significant increase of diffusion rate was observed for samples conditioned at 140°C.

4.2.2. Effect of Temperature on Diffusion

In a more extended work plan, the effect of temperature on binder diffusion was studied. The aim was to determine whether blending and diffusion between fresh and aged binders continues at lower (service) temperatures or not. For this purpose sandwiches of fresh and aged F.H binder were made and conditioned at temperatures ranged from 40°C to 140°C.

For some low conditioning temperatures, samples were conditioned in the DSR and tested instantly after conditioning. Conditioning in the DSR was applicable to low temperatures (40°C and 64°C) because there was no binder bleeding and the geometry could be maintained during the conditioning. The two layered sandwich is shown in Figure 24.



Figure 24 Asphalt binder sandwich conditioning in the DSR

The tests performed with the same specifications as previous binder sandwich and mortar tests. The G^* increase an indication of the blending trend demonstrate the significant effect of temperature on the blending between two binders. Sandwich samples subjected to conditioning temperatures higher than 80°C show blending at different rates (Figure 25).

The two lower conditioning temperatures (40°C and 64°C) show no changes in G^* results for different conditioning times. This indicates that the blending cannot be achieved at low temperatures.

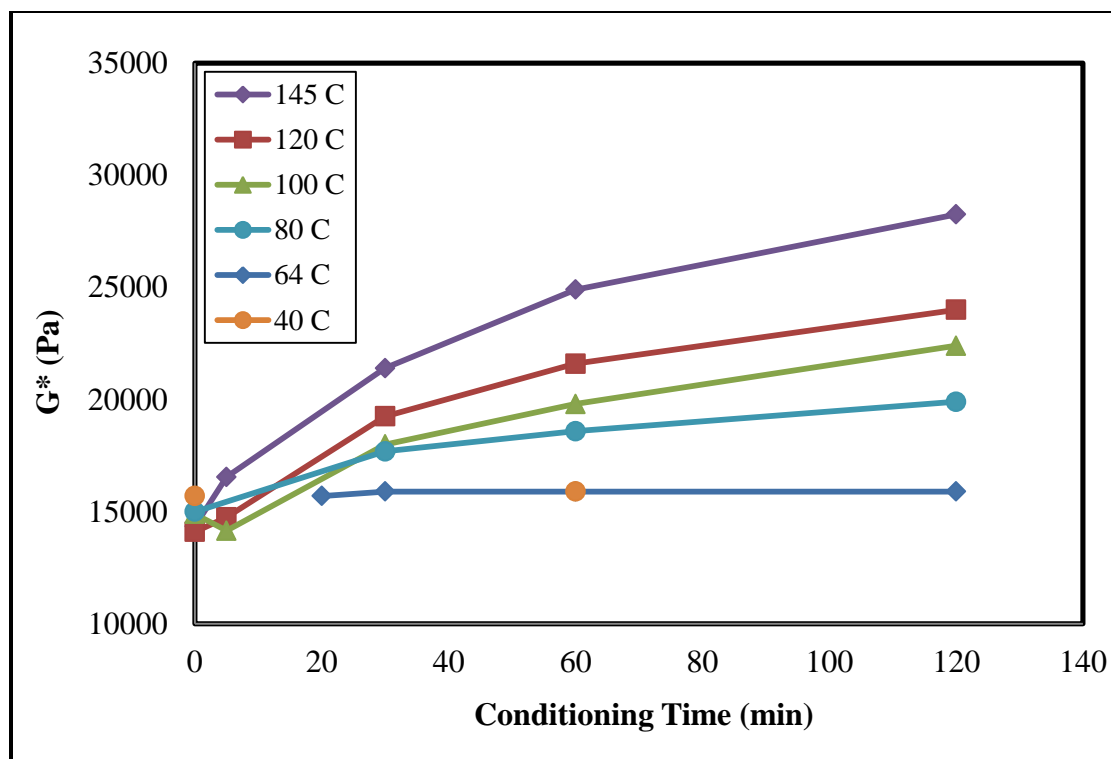


Figure 25 DSR results for FH+Aged FH sandwiches conditioned at different temperatures

Blending rate also can be shown with calculated diffusion coefficient for each temperature. Diffusion coefficients were determined using the experimental measurements and Fick's law solution. Figure 26 exhibits the diffusion coefficients estimated for sandwich samples. Samples conditioned at 145°C show significantly higher diffusion rates while the rates estimated for 100 and 120°C were close. These results were in accordance with calculated diffusion rates for FH+Aged Valero and Valero+Aged FH samples.

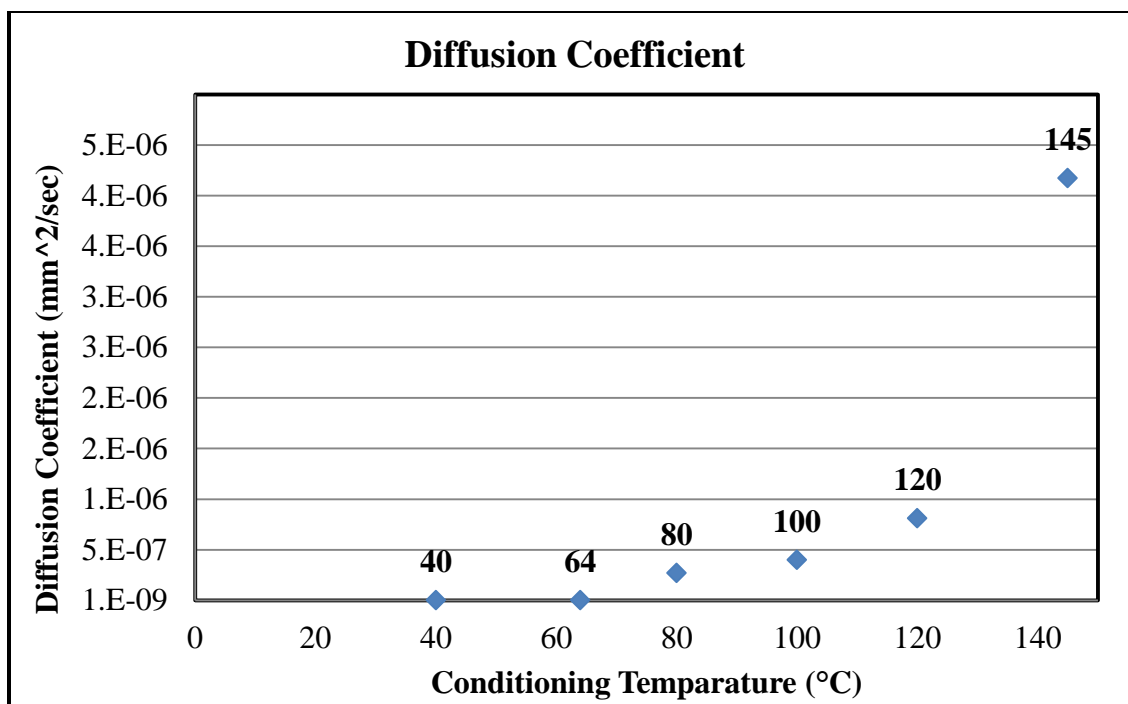


Figure 26 Diffusion coefficients estimated for FH+Aged FH at different temperatures

These results indicate the noticeable effect of temperature on the diffusion phenomena in binders. At a very low temperature the molecular mobility of binders is at a low level and cannot cause noticeable mass transfer, therefore, blending is not expected.

For very low level of molecular mobility, the concentration changes are very small; therefore the diffusion rate for corresponding temperatures cannot be accurately estimated. However, DSR measurements demonstrated no changes for samples conditioned at 40 and 64C which by itself indicates that diffusion does not occur or is not significant enough to change mechanical properties of samples.

4.2.3. Effect of Film Thickness

Another factor which was speculated to affect the diffusion rate was film thickness changes of both fresh and aged binders. Sandwiches of fresh and aged F.H with three film thickness

combinations were made (Table 5). The total sample thickness was 2 mm, to remain consistent with previous DSR testing.

Table 5 Film Thickness Test Setup

Asphalt binders		Thickness		Conditioning	
Fresh	2-PAV	Fresh	2-PAV	Temperature (°C)	Time (min)
FH	FH	1.5	0.5	145	1, 5, 30, 60, 120
		1	1	145	
		0.5	1.5	145	

DSR tests were performed with 2mm gap at 64°C. The measured G^* s were normalized to the initial G^* of the relevant sample set. Figure 27 shows the G^*/G^*_{initial} for asphalt binder sandwiches at different temperatures and film thickness combinations.

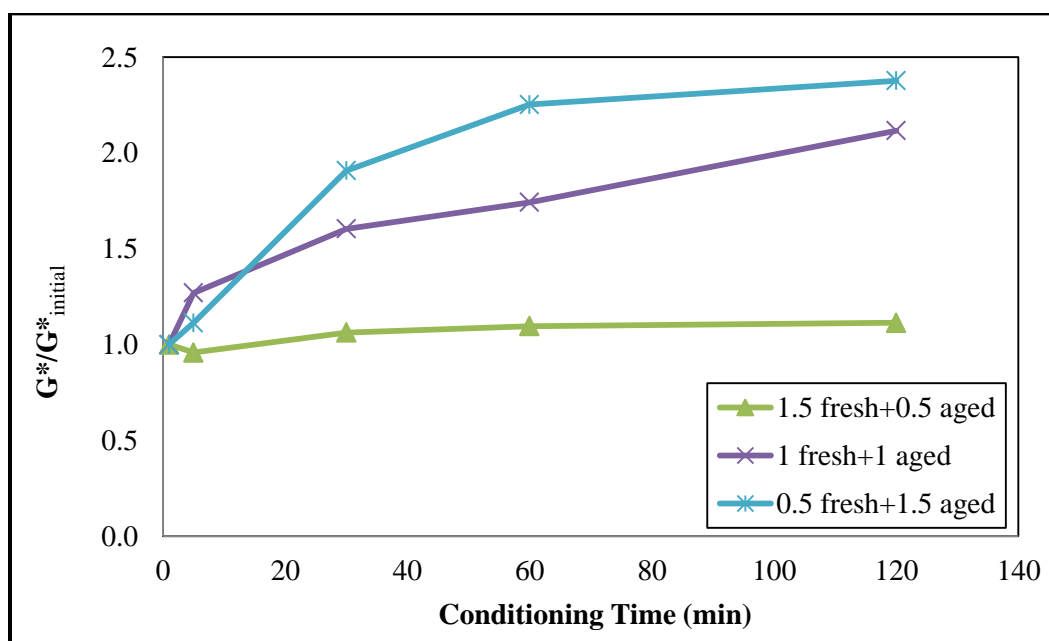


Figure 27 Normalized G^* results for different thickness combinations conditioned at 145°C

Demonstrated in the Figure 27, a higher rate of G^* increase was obtained for binder sandwiches with higher thickness of aged binder or lower thickness of fresh binder. The trend for sample with 0.5 mm of aged binder exhibited very small changes with conditioning at 145°C.

Although, Figure 27 provides a trend to determine the effect of film thickness, the results may be confounded by the higher sensitivity of composite G^* to the thickness of the softer medium (fresh binder). Figure 28 Provides a theoretical analysis (based on applied composite theory) of dependency of total G^* on the thickness of the fresh binder for a total 2mm thick sandwich sample used in experiments.

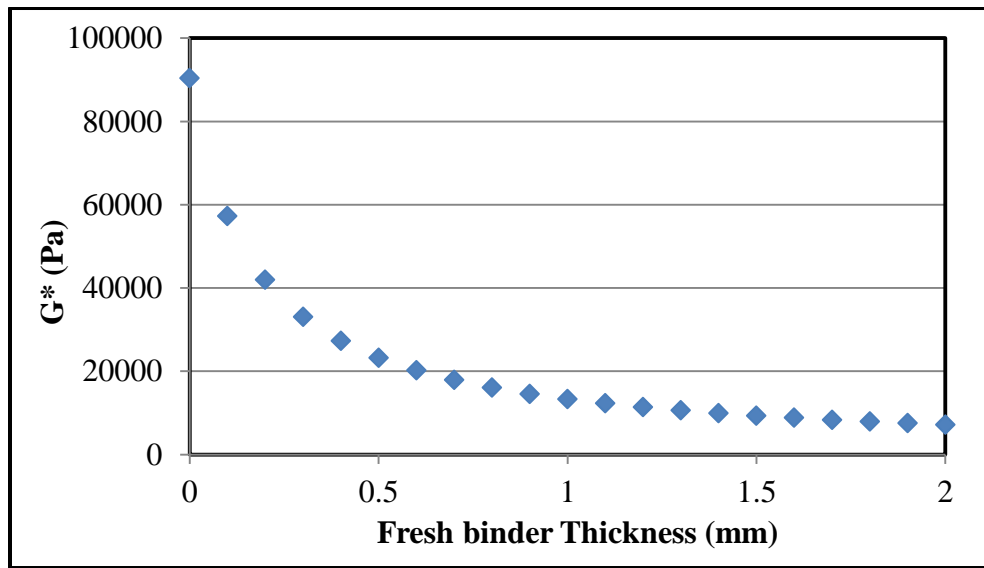
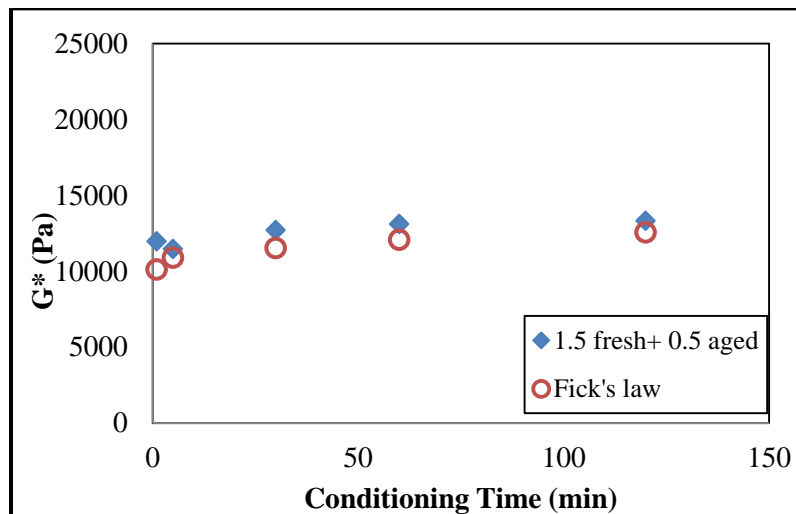


Figure 28 Effect of fresh binder thickness in a 2mm binder sandwich (fresh + aged binder)

Based on this analysis, at lower thicknesses of fresh binder, higher composite G^* changes is expected while for higher thicknesses the effect of change in fresh binder thickness is minor. Therefore, a comprehensive conclusion cannot be made about the film thickness effect using this test method.

Based on the experimental measurements, it was tried to fit Fick's law calculations and determine the diffusion coefficient for combinations. The applied model still was capable to fit theoretical results to the experimental data for fresh binder thicknesses larger or equal to aged binder (thickness of fresh binder \geq thickness of aged binder). However, for aged binder thickness of larger than fresh binder thickness, acceptable correlation was not obtained (Figure 29). In addition this condition is not likely because in most cases in practice the allowed aged binder is usually much less than the fresh binder. The condition shown in Figure 28 deserves further research.



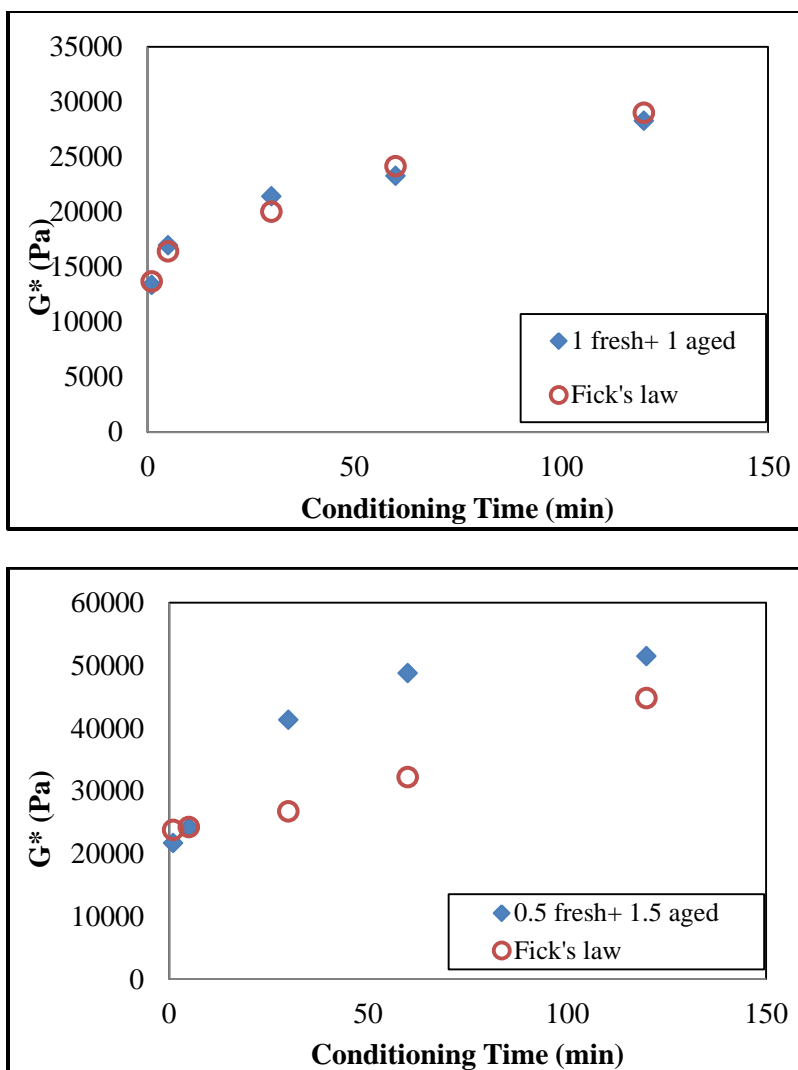


Figure 29 effect of binders' thickness based on Fick's law correlations

According to the Fick's law theory used in this study (assumption of no concentration dependency) the diffusion rate is constant for a conditioning temperature and independent from initial thickness of fresh and aged binders. Therefore, in a thin interface section of all three configurations, the same diffusion coefficient should be obtained for a determined conditioning temperature. The blending extension occurs at faster rates at shorter time of conditioning and it is highly probable that the small thickness of fresh binder is blended with the aged binder and the assumption of concentration independency no longer holds while for higher thicknesses, the

blending still can develop in the ranges of applied conditioning time. This means that the applied Fick's law model cannot give a good correlation for long conditioning time of samples with small fresh binder thickness and is concentration dependent.

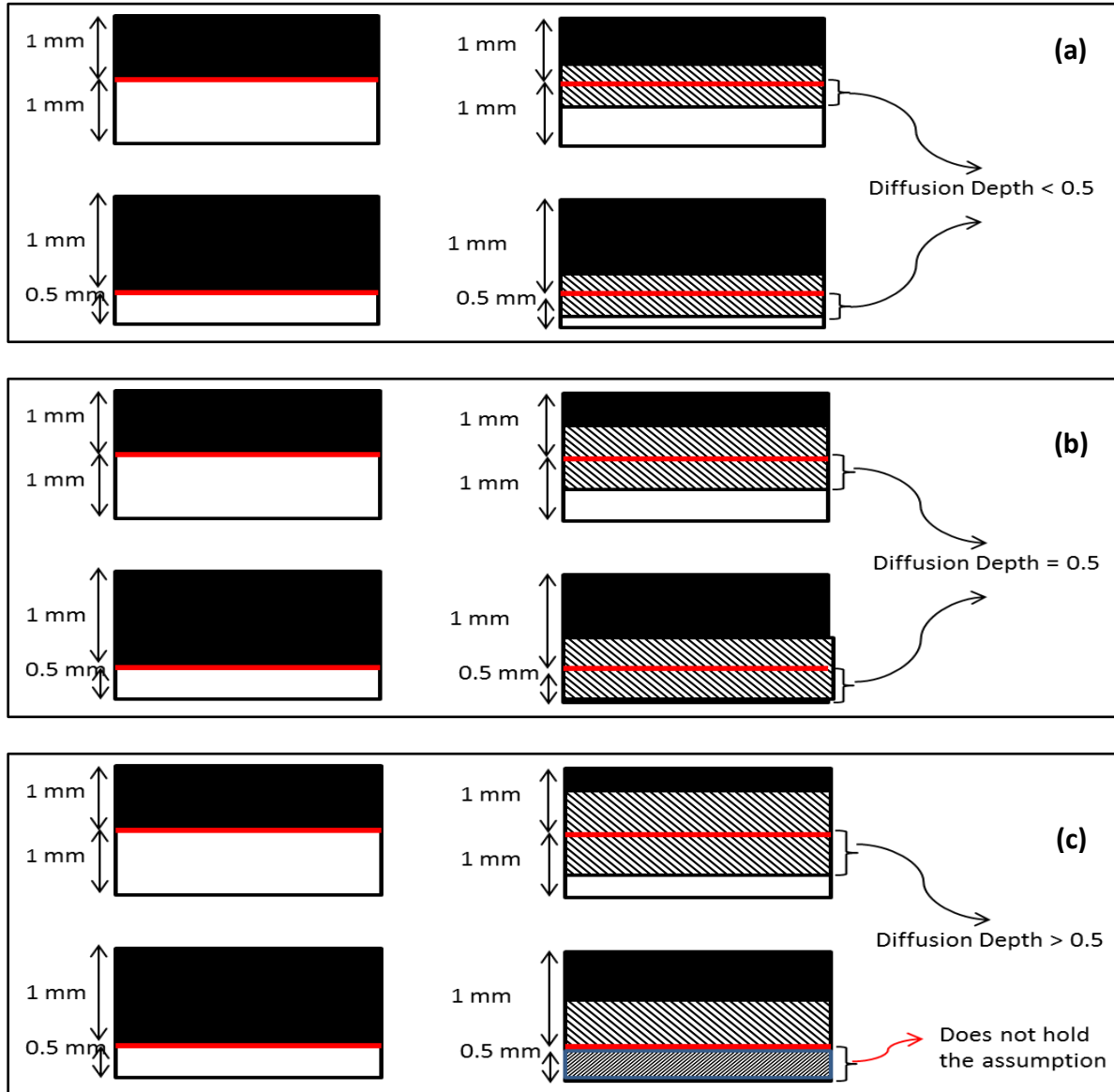


Figure 30 Schematic of blending extension compared for two combinations; for diffusion depth higher than the thickness of fresh binder, the boundary condition and assumption of concentration independency does not hold

In addition this condition is not likely because in most cases in practice the allowed aged binder is usually much less than the fresh binder. The condition shown in Figure 28 deserves further research.

5. Findings, Conclusions, and Recommendations

5.1. Main Findings

A testing and analysis procedure was developed and proposed in this study to estimate the level of blending between fresh and RAP binders as a function of conditioning time and temperature. The blending between two binders occurs if they are in contact at proper temperatures for enough time. The procedure includes using two binder samples placed over each other and tested in the DSR. The G^* results measured by the DSR for the 2-layer sample increases with conditioning time at high enough temperatures indicating the effect of blending. These results are shown to follow the applied diffusion law (Fick's law) and a constant diffusion coefficient can be determined by fitting the estimated and measured G^* for a combination of two binders.

The level of blending measured using the DSR two-layer procedure is also verified using RAP mortars consisting of fresh binder mixed with selected sizes of RAP material. Results of mortars and binder two-layer samples indicate that conditioning temperature has a significant effect on the blending trend and the final achievable blending level. The diffusion coefficients also demonstrated a considerable dependence on conditioning temperature.

5.2. Summary of Findings and Conclusions

- The binder two-layer method in the DSR is suitable for determining the blending between fresh and aged binders. The sample preparation is easy and applicable. Test results using the DSR were very repeatable.

- Using DSR measurements, the diffusion coefficient can be calculated for two binders in contact. The diffusion coefficient is constant with time, but increases with temperature, due to the high dependence on the viscosity of the binders. This finding agrees with previous studies on the diffusion mechanism.
- In addition to viscosity there are other factors that contribute to diffusion development. More research is needed to identify these factors.
- The diffusion phenomenon was confirmed by using actual RAP mortars, as well as artificial RAP mortars.
- In order to achieve full blending of fresh and aged binders in recycled asphalt mixtures, the temperature of conditioning should be specified and controlled in the field. In this study it is shown that at conditioning temperatures below 100°C (corresponding to viscosities higher than 3 Pa.s) very limited blending between the fresh and RAP binder occurs, thus these conditions should be avoided in use of RAP in pavements.

5.3. Recommendations

- It is recommended that the results of this study to be extended and validated using binders extracted from field produced RAP mortars and mixtures.
- The effect of polymer and Warm Mix modifiers on diffusion and blending of fresh and RAP material must be investigated.
- Furthermore, in addition to the Dynamic Shear Rheometer methodology used in this study, other evaluation methods such as FTIR, or detectable pigments in the binder, can be used to assess the blending and diffusion development between fresh and RAP binders.

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