

### **3.5 Routes to durability and sustainability: Recycling of PmB containing RAP**

*Dr. Xueyan Liu, Associate professor*

*Delft University of Technology, The Netherlands*

#### **Abstract**

Recycling of base asphalt pavement with rejuvenators or soft binders has been applied for many decades and a lot of experience has been gained. However, the high-quality recycling technique of polymer modified asphalt (PMA) has still not been sufficiently studied. The aging mechanism of PMA is complex, including the combined effects of the oxidation of bitumen and the degradation of the polymer. The current commercially available rejuvenators are designed mostly for base bitumen and not suitable for the recycling of PMA. For this reason, this research aims at designing an innovative rejuvenator specifically for the recycling of PMA. Firstly, a series of performance-based test methods, including viscoelastic properties, rutting resistance, fatigue resistance, cracking resistance, relaxation ability and aging ability, has been performed to select the appropriate source materials and to determine the optimum ratio between different components. After that, a specific SBS-based rejuvenator was found to be most effective in PMA rejuvenation. To reveal the rejuvenation mechanism, an environment scanning electron microscope (E-SEM) was utilized in investigating the microstructure of the rejuvenated binder. The results illustrated that E-SEM method can distinguish the influence of rejuvenator dosage, rejuvenator types, and addition of fresh bitumen on the morphology of aged PMB, which can help us to have a better understanding of the rejuvenation mechanism.

#### **About the speaker**



**Dr. Xueyan Liu** is currently an associate Professor in the Section of Pavement Engineering of the Faculty of Civil Engineering & Geosciences of TU Delft. He works in the areas of constitutive modelling, numerical modelling and material experimental characterization. Within the research program of the Section Pavement Engineering, his research topics mostly relate to the development and implementation of constitutive models for the simulation of the static and dynamic response of various pavement engineering materials like soils, asphalt concrete, liner and reinforcing systems etc. and sustainable development technologies, i.e., multiscale modelling of asphaltic materials, warm/cold asphalt concrete technology, durability of asphalt surfacings on orthotropic steel deck bridge, accelerated pavement test, pavement continuous monitoring and sustainable development technologies. Dr. Liu was granted his doctoral thesis in 2003. During the same period, Dr. Liu participated also in the team that developed the ACRe model for Asphalt Concrete Response currently implemented in 3D Computer Aided Pavement Analysis system (CAPA-3D).

Dr. Liu has published more than 100 technical and journal papers on the mechanics and the finite element modelling of granular, concrete and asphaltic materials. Dr. Liu is a member of RILEM Technical Committee 272-PIM Phase and Interphase behaviour of Bituminous Materials and a member of Delft Centre for Materials (DCMat). He is also a member of ISAP, AAPT, APSE and IACMAG. Dr. Liu is an Editorial Board Member of Geomaterials (GM). Dr. Liu was appointed as Board member of the International Association of Chinese Infrastructure Professionals (IACIP) and member of the Academic Committee of the Key Laboratory of Road Structure and Materials Transportation Industry of the China Ministry of Transport. He is also actively involved in organizing inter/national workshops and conferences and was invited as Scientific/Technical committee member of several international conferences.



**5th International Symposium  
on Frontiers of Road and Airport Engineering**

Workshop: Changes in binder properties and the role of additives  
**Routes to durability and sustainability: Recycling of PmB containing RAF**

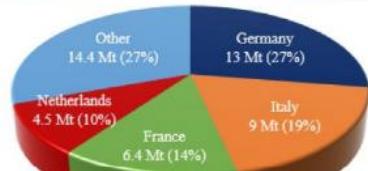
Xueyan Liu, Peng Lin, Sandra Erkens  
Delft University of Technology, the Netherlands



**TU**Delft



# 1. Introduction- Recycling



- In Europe, 49.44 mt RA available (figures 2018)
  - 68% reused into new Asphalt mixture,
  - 19% recycled as granular materials in unbound layers
- In China, 117 mt RA available
- In the US, 91.7 mt RA available
- In Japan, 37 mt RA available

# 1. Introduction- Recycling

## European Vision

- Sustainability.
  - Reduction of CO2 emission, 80% by 2050 (1990 baseline)
- Circular economy\*,
  - Dec 2015 action plan → implemented in March 2019
  - Avoid, Reduce, Reuse, Recycle, Treat, Dispose along the whole life cycle of products
- Green Product Procurement\*\*,
  - Integrating circular economy requirements
  - 2016, criteria for road design, construction and maintenance

\*[http://ec.europa.eu/environment/circular-economy/index\\_en.htm](http://ec.europa.eu/environment/circular-economy/index_en.htm)

\*\* [http://ec.europa.eu/environment/gpp/pdf/report\\_gpp\\_office\\_buildings.pdf](http://ec.europa.eu/environment/gpp/pdf/report_gpp_office_buildings.pdf)



## 1. Introduction- Recycling



### Asphalt Recycling in the Netherlands

- 71% of the RAP is used in HMA and WMA recycling
- 11% of the RAP is used in Cold recycling
- 18% RAP is used in other applications.

### New goals In the Netherlands

- 50% CO2 and raw material reduction in 2030
- Be circular ( no waste, no use of raw materials) in 2050

## 1. Introduction- Marketing



### PMB Market Distribution



### PMB in Global Market

**More than 80% of PMB was used in pavement construction.**

*Global Polymer Modified Bitumen Market Professional Survey Report 2018*

# 1. Introduction- Challenge



EAPA Workshop

Use of rejuvenators in asphalt mixtures  
Padova (Italy), 10th - 11th September 2019

## 1. INTRODUCTION

### Now

The reuse of RAP with standard bituminous binders up to 50 % addition rates is successfully achieved.

### Challenges

Over recent years more **PMB** has been used in asphalt production. However, 'standard technology' may not be adequate for recycling such kind of RAP.

... and improves performance under many conditions. Furthermore, rejuvenating additives restore the rheological behaviour of the aged binder from reclaimed asphalt when blended with this (pre-heated) reclaimed asphalt. Rejuvenators also can avoid the need of using a soft paving grade bitumen to meet the requirements.

# 1. Introduction- Goal



## High-quality Recycling of PMB-RAP



### Improve Quality

- **Activate** the aged PMB in the RAP
- **Fully use** the remaining polymer
- **Longer service life** of pavement



### Increase Profit

- **Increase** the PMB-RAP content
- **Societal** benefits
- **Decrease** the release of **CO<sub>2</sub>**

# 1. Introduction- Research Scheme

## Recipe Optimization



- Material Selection
- Formula Optimization



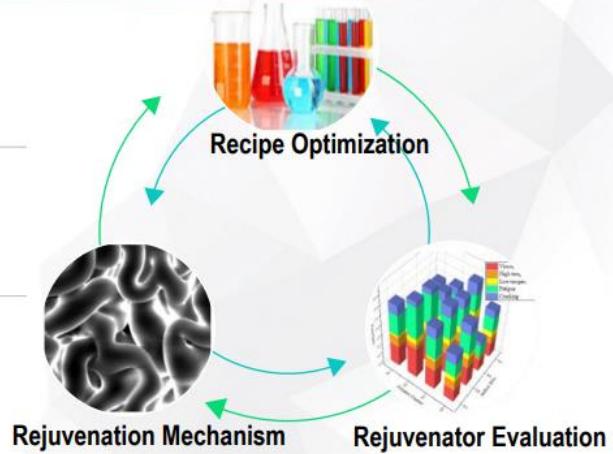
## Rejuvenator Evaluation

- Rheological Evaluation
- Rejuvenator Dosage Determination
- Multi-cycles Recycling Evaluation



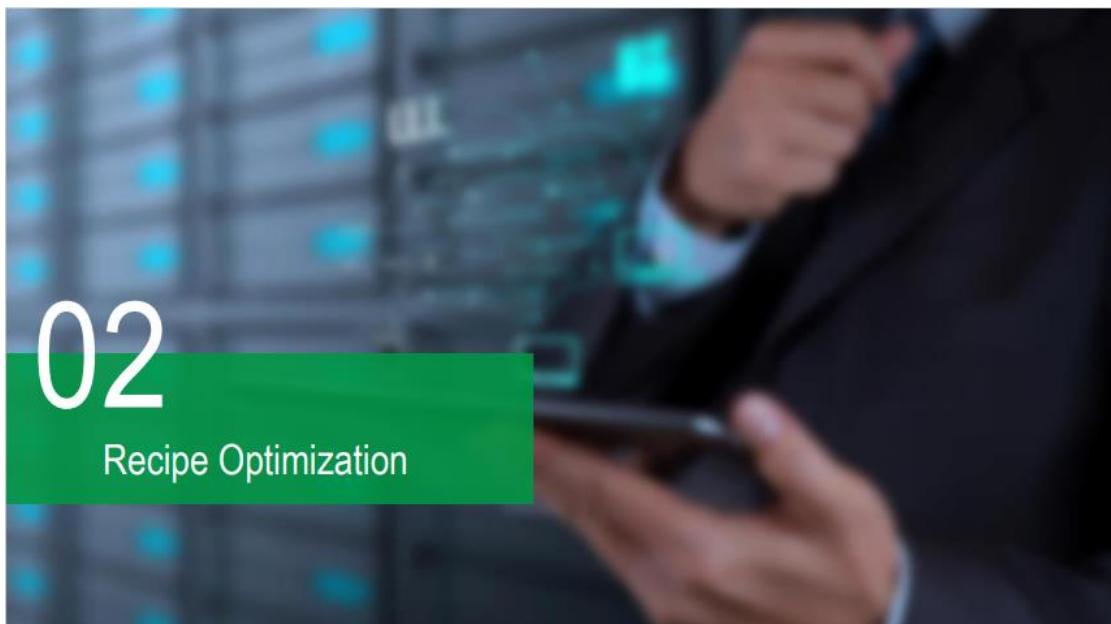
## Rejuvenation Mechanism

- Colloidal Stability
- E-SEM micro-structure
- DSC thermal analysis



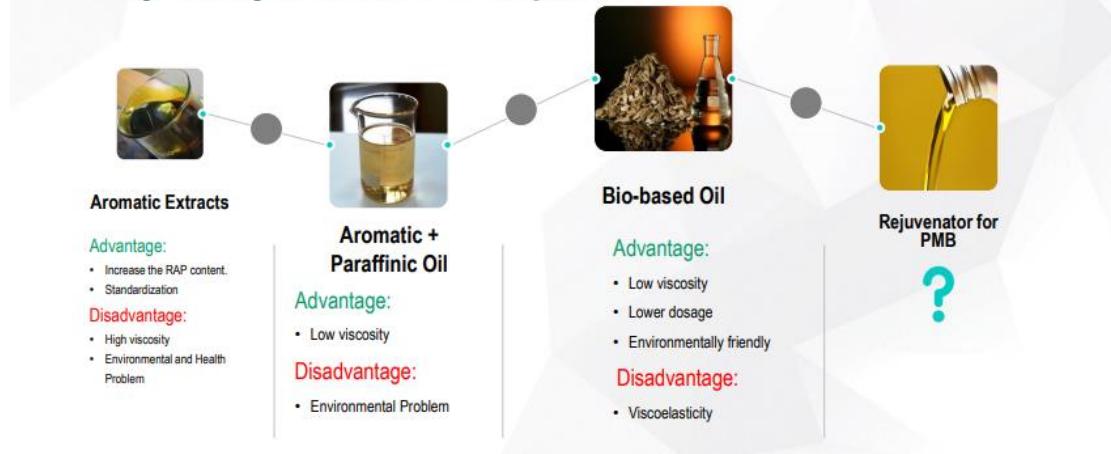
02

## Recipe Optimization



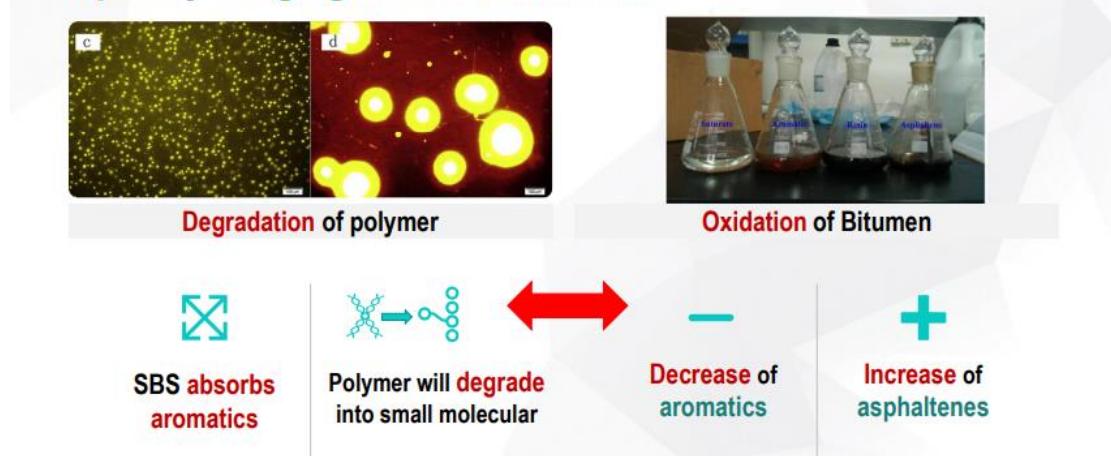
## 2.1 Recipe Optimization

### History of Rejuvenator Development



## 2.1 Recipe Optimization

### Complexity of aging mechanism in PMB



## 2.1 Recipe Optimization

### Challenges in rejuvenation of PMB

1. Hardening due to the oxidation of base bitumen

2. Property loss due to the degradation of polymer

3. Colloidal stability loss in the recycled PMB

## 2.1 Recipe Optimization

### Procedure of recipe optimization



Materials Selection



Determination of component ratio in rejuvenator



Mechanical property characterization

## Material Selection



03

Rejuvenator Evaluation

# Research Scheme

## Recipe Optimization



- Material Selection
- Formula Optimization

## Rejuvenator Evaluation

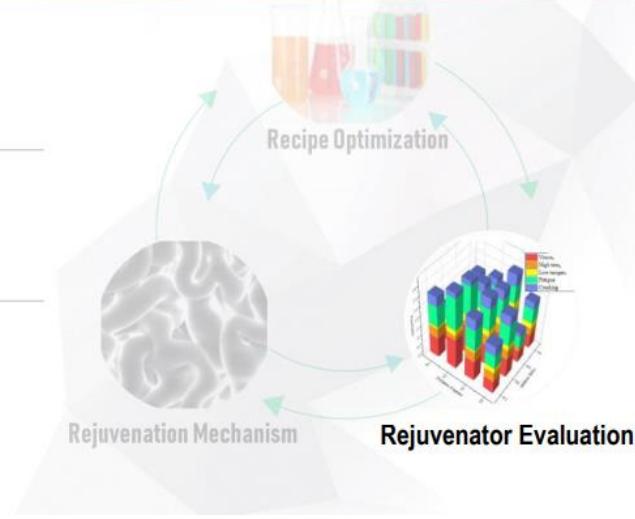


- Rheological Evaluation
- Rejuvenator Dosage Determination
- Multi-cycles Recycling Evaluation

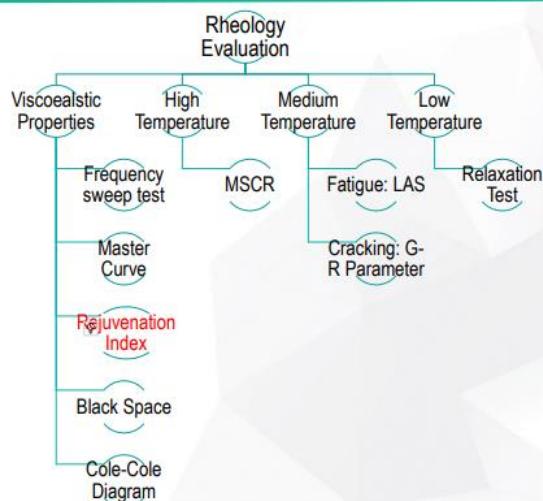
## Rejuvenation Mechanism



- Colloidal Stability
- E-SEM micro-structure
- DSC thermal analysis



## 3. Rejuvenator Evaluation- Rheological Properties



### 3. Rejuvenator Evaluation- Rheological Property TU Delft Delft University of Technology

#### Rejuvenation Indexes

$$A_{MII} = \int_0^4 \log G^*(\xi) d\xi$$

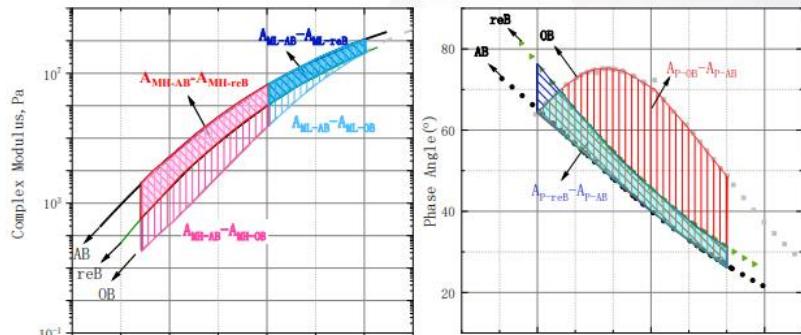
$$A_{MI} = \int_{-5}^0 \log G^*(\xi) d\xi$$

$$A_P = \int_{-5}^4 \delta(\xi) d\xi$$

$$I_{MII} = \frac{A_{MII-AB} - A_{MII-reB}}{A_{MII-AB} - A_{MII-OB}} \times 100\%$$

$$I_{MI} = \frac{A_{MI-AB} - A_{MI-reB}}{A_{MI-AB} - A_{MI-OB}} \times 100\%$$

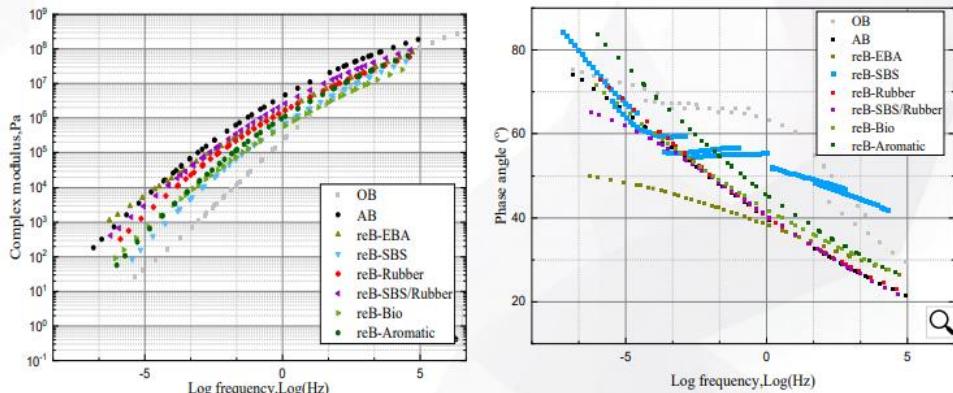
$$I_P = \frac{A_{P-reB} - A_{P-AB}}{A_{P-OB} - A_{P-AB}} \times 100\%$$



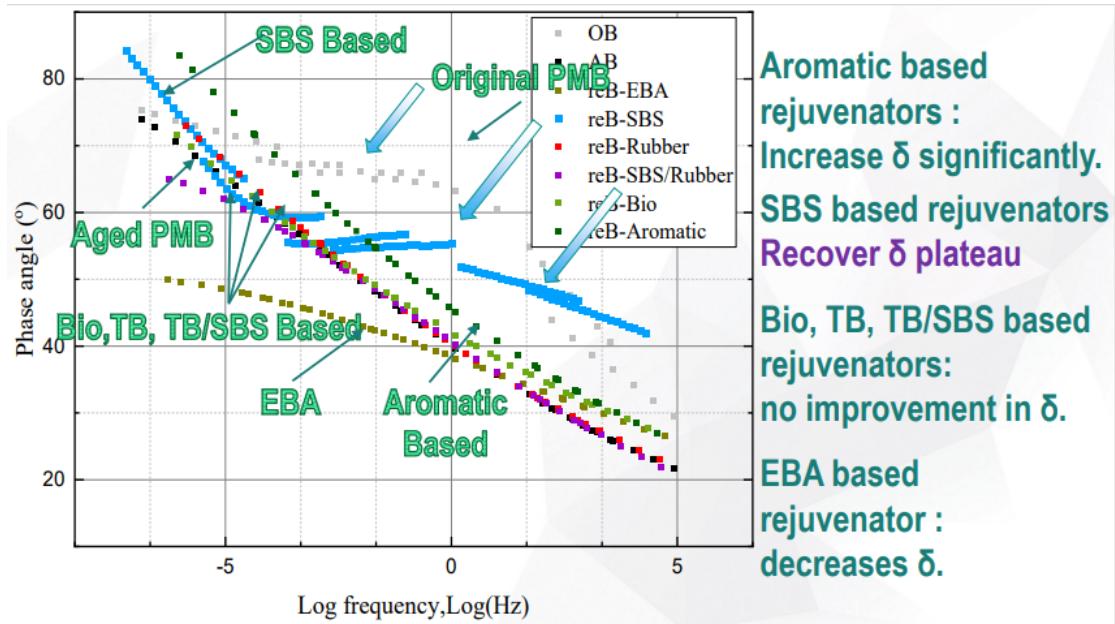
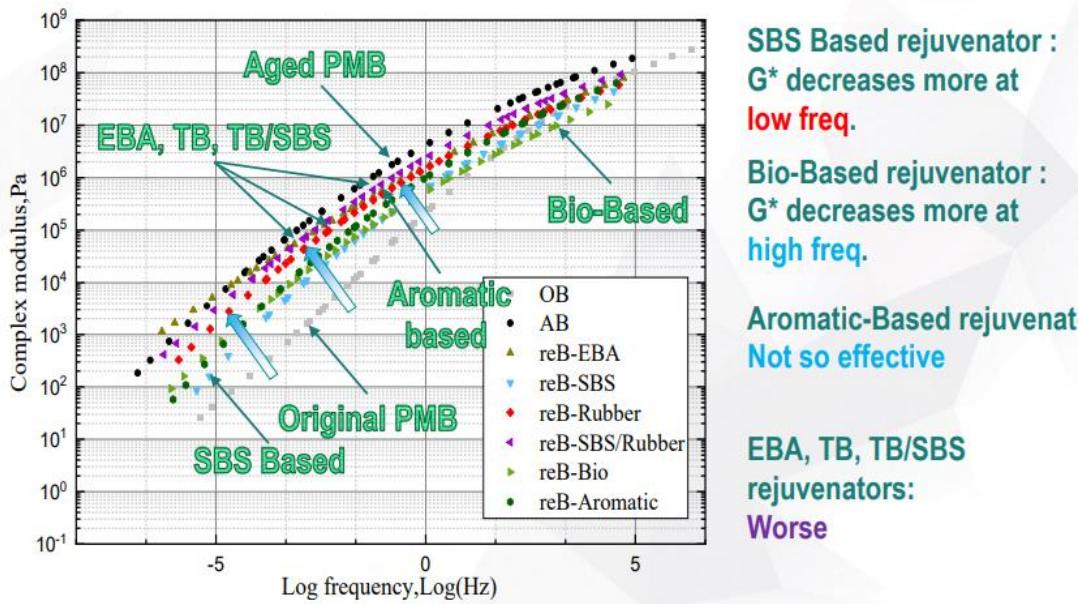
Indexes were defined as the ratio of the integral area difference of  $G^*$  &  $\delta$  between aged and rejuvenated bitumen in the selected frequency range

### 3. Rejuvenator Evaluation- Rheological Property TU Delft Delft University of Technology

#### Viscoelastic-Master Curves

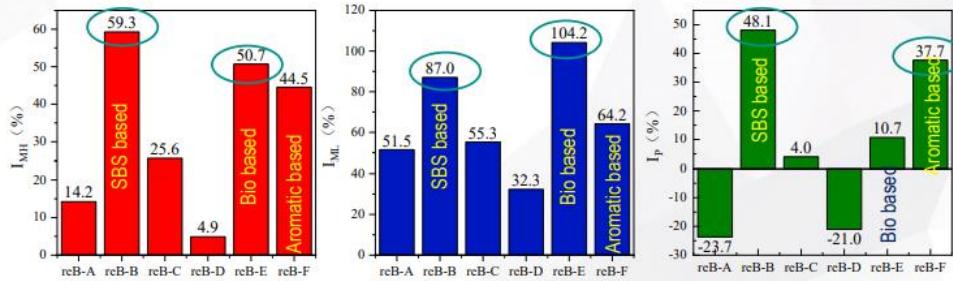


DSR frequency sweep tests between 0.01 and 10 Hz were performed at 0, 20, 30, 40, 60 and 80°C  
the reference temperature is 30 °C.



### 3. Rejuvenator Evaluation- Rheological Property Delft University of Technology

#### Rejuvenation Index



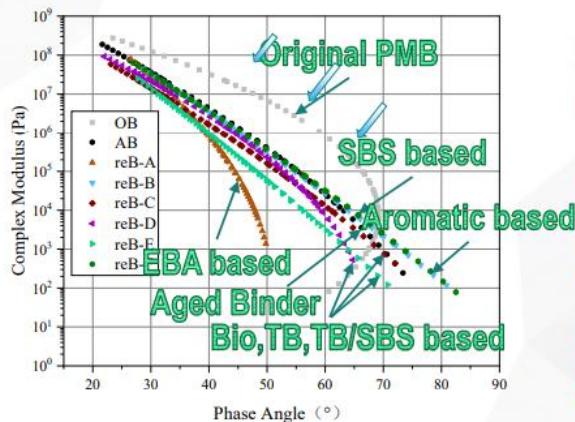
$$I_{MH} = \frac{A_{MH-AB} - A_{MH-reB}}{A_{MH-AB} - A_{MH-OB}} \times 100\%$$

$$I_{ML} = \frac{A_{ML-AB} - A_{ML-reB}}{A_{ML-AB} - A_{ML-OB}} \times 100\%$$

$$I_P = \frac{A_{P-reB} - A_{P-AB}}{A_{P-OB} - A_{P-AB}} \times 100\%$$

### 3. Rejuvenator Evaluation- Rheological Property Delft University of Technology

#### Viscoelastic-Black Space



Influence on viscoelastic property:

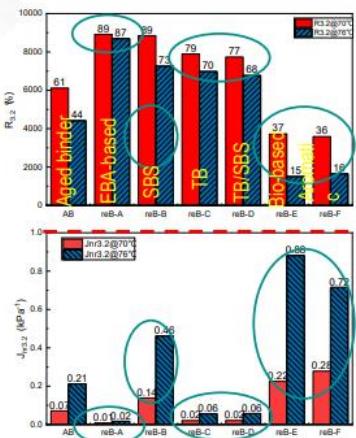
**SBS and Aromatic based rejuvenators have positive effect**

**Bio, TB and TB/SBS based rejuvenators have a certain adverse effect**

**EBA based rejuvenator has significant adverse effect**

### 3. Rejuvenator Evaluation- Rheological Property

#### High Temperature Properties



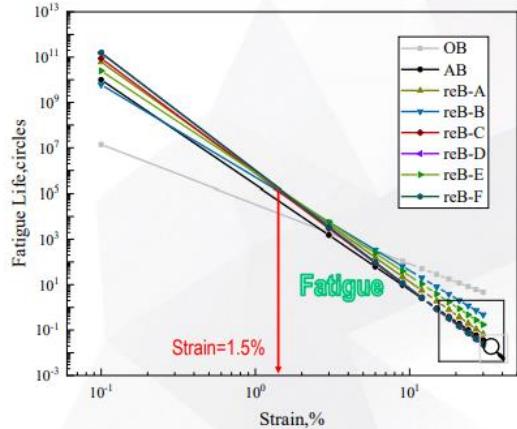
- EBA, TB, TB/SBS based rejuvenators have advantages in MSCR results.
- SBS based rejuvenator shows better strain recovery capacity compared with the reference rejuvenators.
- $J_{nr3.2}$  values of all binders at 76°C are much lower than the maximum allowable value ( 2  $kPa^{-1}$  ) for the 'H' traffic level.

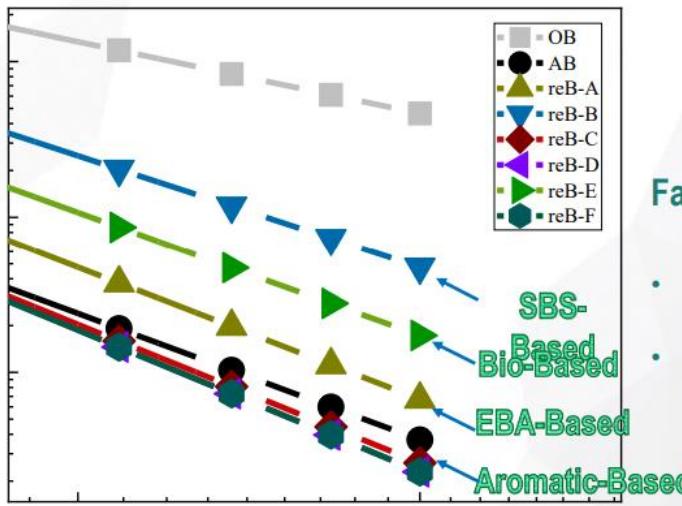
### 3. Rejuvenator Evaluation- Rheological Property

#### Medium Temperature-Fatigue



LAS Test @ 10Hz & 20 °C



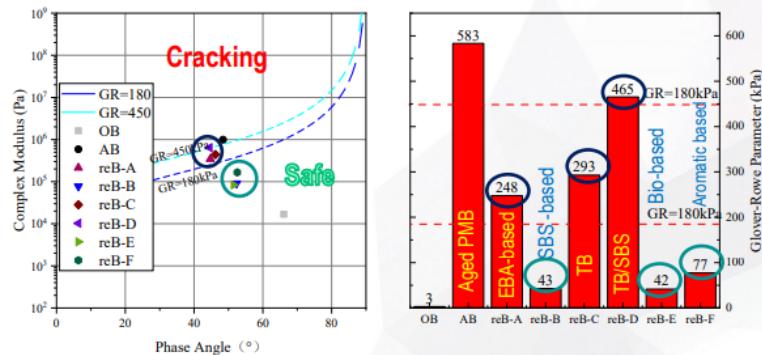


### Fatigue resistance :

- SBS based > Bio-based > EBA base
- Aromatic, TB, TB/SBS have **adverse effect**

## 3. Rejuvenator Evaluation- Rheological Property Delft University of Technology

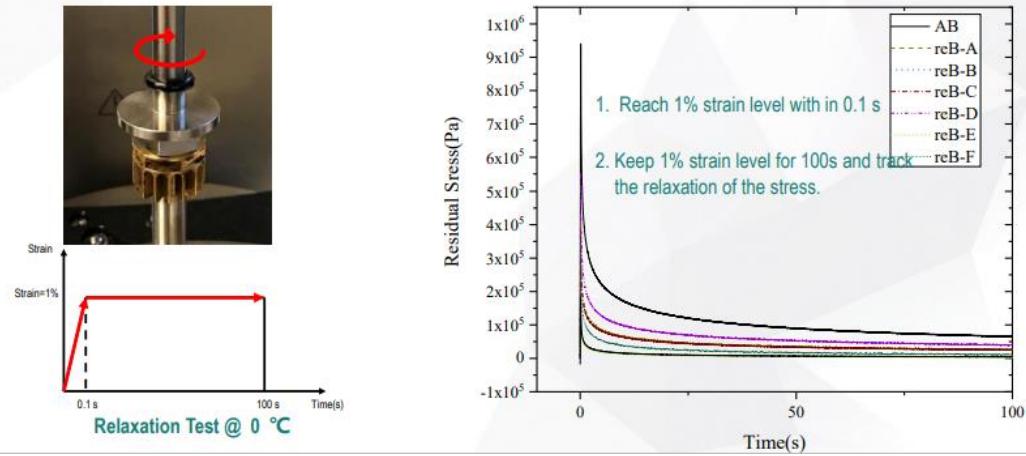
### Medium Temperature-Cracking evaluation with G-R parameter



**SBS, Bio, Aromatic based rejuvenators significantly improve cracking resistance**

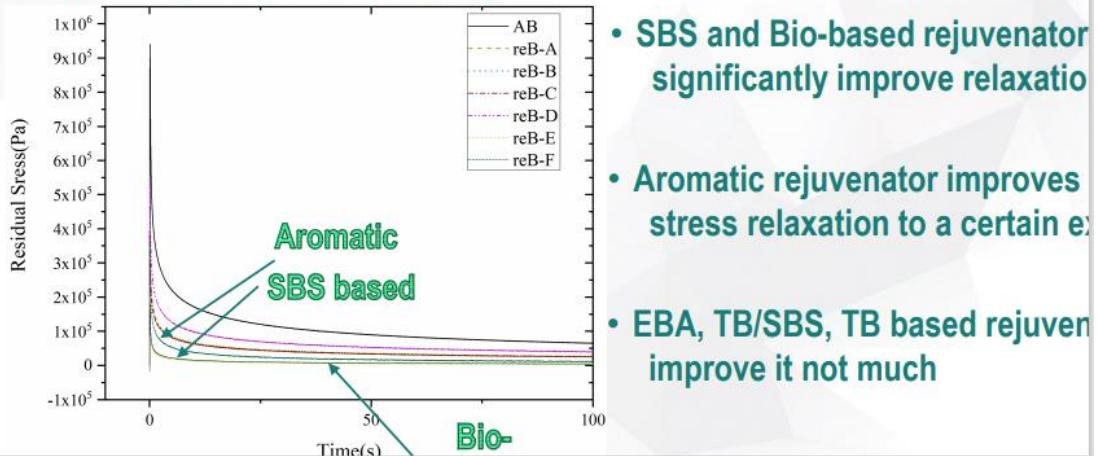
### 3. Rejuvenator Evaluation- Rheological Property

#### Low Temperature-Relaxation Test

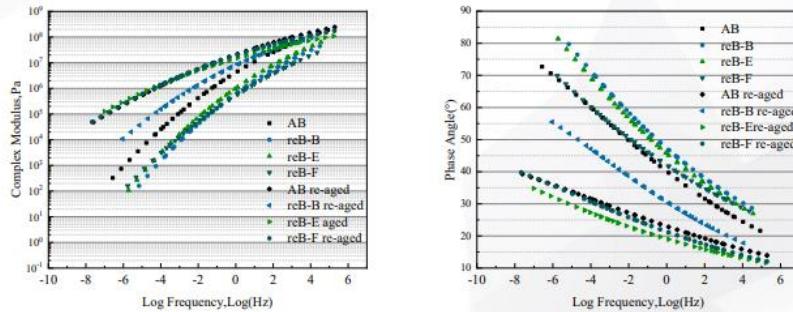


### 3. Rejuvenator Evaluation- Rheological Property

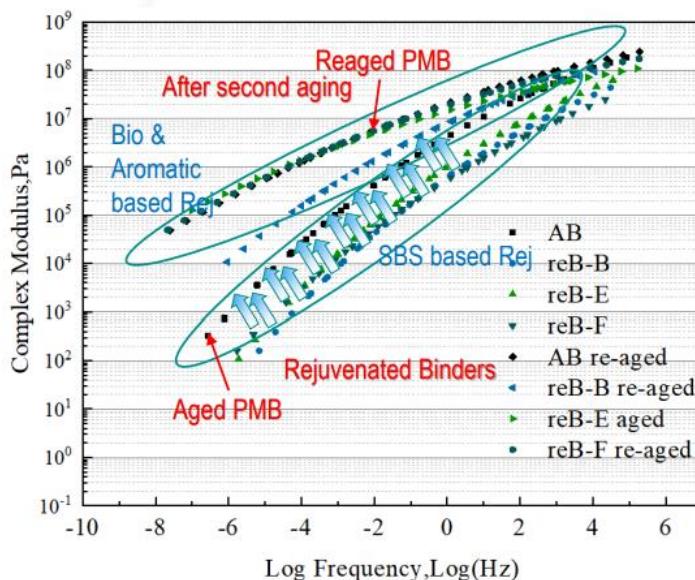
#### Low Temperature-Cracking



## Anti-aging Properties



Xueyan Liu, Peng Lin et.al. "Effect of Rejuvenators on Performance-based Properties of Aged Polymer Modified Bitumen" .99<sup>th</sup> Transportation Research Board.

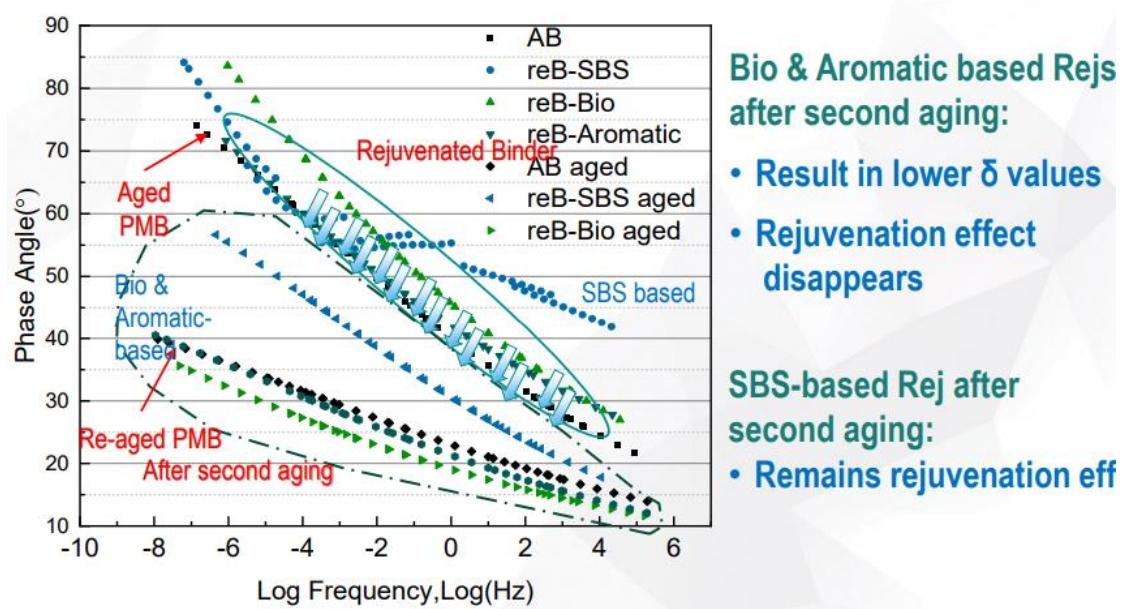


### Bio & Aromatic based rejuvenators after second aging:

- $G^*$  values are almost same
- Rejuvenation effect disappears

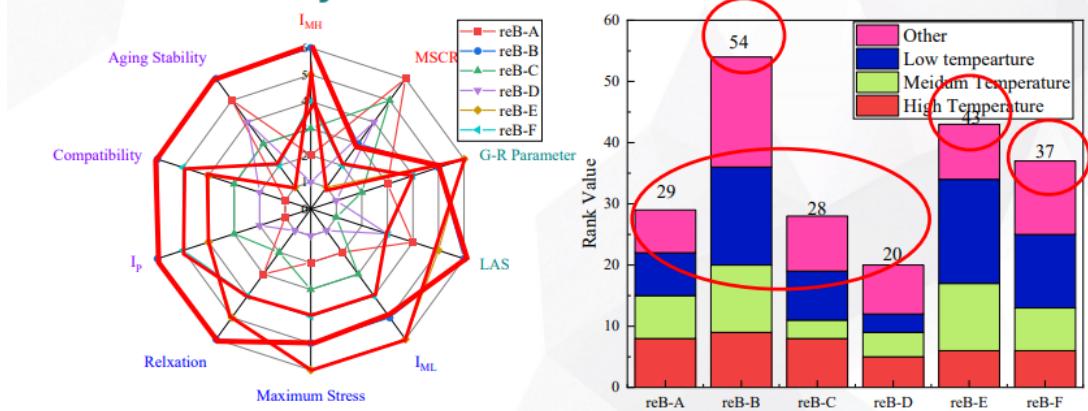
### SBS-based rejuvenator after second aging:

- Remains rejuvenation effect



### 3. Rejuvenator Evaluation- Rheological Property Delft University of Technology

#### Radar Chart Analysis



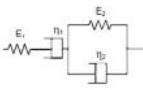
# Research Scheme of Rejuvenator



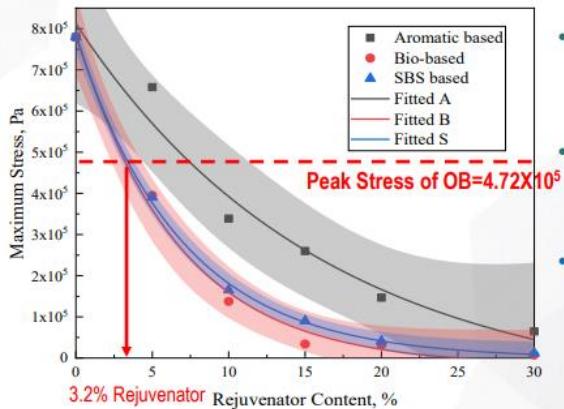
## 3. Rejuvenator Evaluation- Dosage Determination

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### Performance based methods

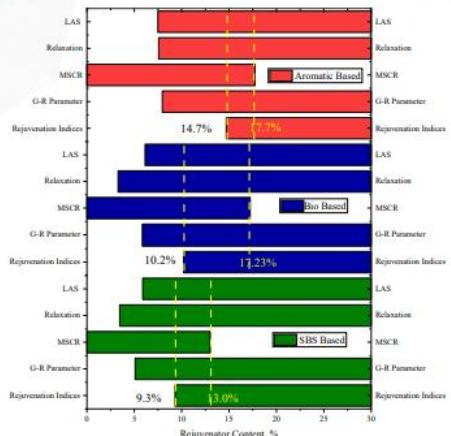
Viscoelastic Properties	High Temperature	Medium Temperature	Low Temperature
 <ul style="list-style-type: none"> <li>Frequency sweep test</li> <li>Rejuvenation Efficiency</li> <li>Master curves</li> <li>Black Space Cole-Cole Diagram</li> </ul>	 <ul style="list-style-type: none"> <li>MSCR</li> <li>Rutting Parameter</li> </ul>	 <ul style="list-style-type: none"> <li>Fatigue: LAS</li> <li>Cracking: G-R</li> </ul>	 <ul style="list-style-type: none"> <li>Relaxation Test</li> </ul>

## Low Temperature- Relaxation



- The maximum stress change shows an exponential relationship with rejuvenator dosage,
- SBS and Bio-based rejuvenators are more efficient than Aromatic-based rejuvenator in decreasing maximum stress
- Calculate the minimum rejuvenator dosage based on the peak stress equals to the one occurred in the original fresh binder

## Rejuvenator Dosage Determination

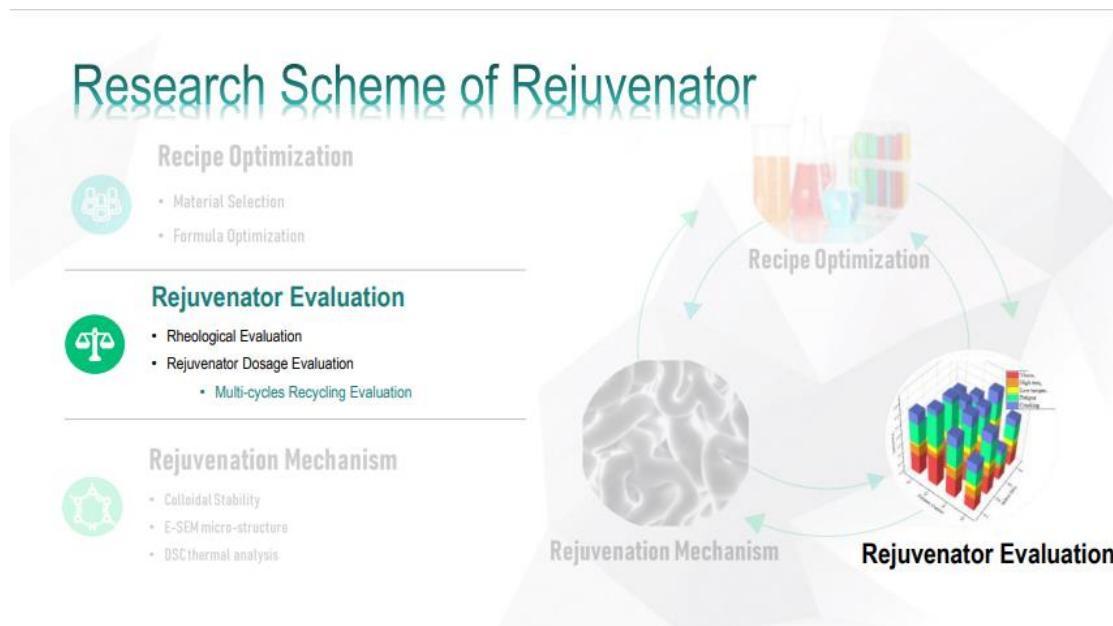


Traditional Methods  
(Penetration, Softening point)

Performance Based  
Methods  
(High, Low, Medium  
performance)

Peng Lin, Xueyan Liu, Panos Apostolidis, Sandra Erkens, Shisong Ren, Shi Xu, Tom Scarpas, and Weidong Huang. "On the Rejuvenator Dosage Optimization for Aged SBS Modified Bitumen." *Construction and Building Materials* 271 (February 15, 2021): 121913.

# Research Scheme of Rejuvenator

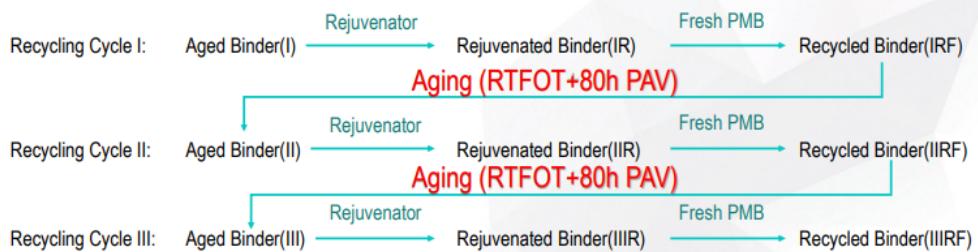


## 3. Rejuvenator Evaluation- Multi-Cycles Recycling

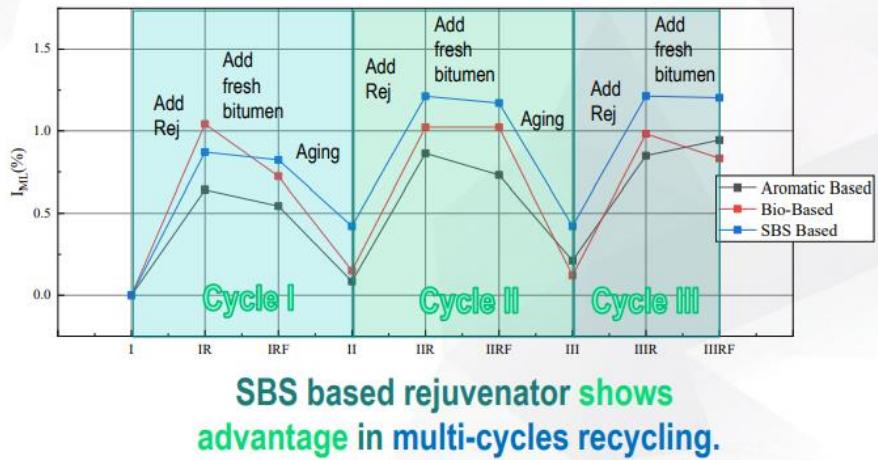
Delft University of Technology



1. Is the rejuvenator still functional?
2. How are the rheological properties influenced?
3. ....



### 3. Rejuvenator Evaluation- Multi-Cycles Recycling



SBS based rejuvenator shows advantage in multi-cycles recycling.

04

Rejuvenation Mechanism

# Research Scheme

## Recipe Optimization



- Material Selection
- Formula Optimization

## Rejuvenator Evaluation



- Rheological Evaluation
- Rejuvenator Dosage Evaluation
- Multi-circle Recycling Ability Evaluation

## Rejuvenation Mechanism



- Chemical analysis
- E-SEM micro-structure analysis
- DSC thermal analysis

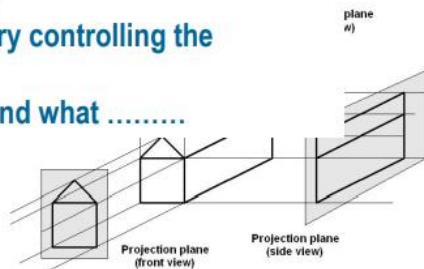


## 4. Rejuvenation Mechanism

- how is the compatibility of rejuvenator with the aged bitumen?
- how is the chemistry controlling the rheology?
- answer how, why and what .....



E-SEM  
Micro-  
structure  
Compatibility



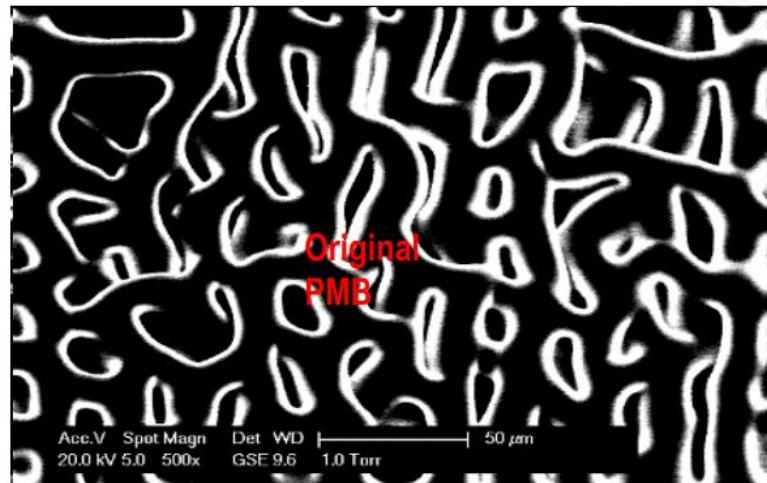
DSC  
Calorimetry  
State of Crystallization



## 4. Rejuvenation Mechanism

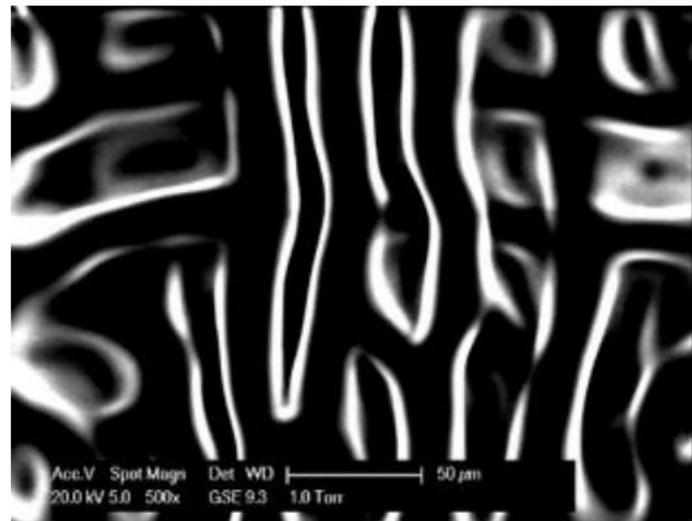


Lin, Peng, X. Liu, P. Apostolidis, S. Erkens, Y. Zhang, and S. Ren. "ESEM Observation and Rheological Analysis of Rejuvenated SBS Modified Bitumen." *Materials & Design* 204 (June 1, 2021): 109639.



“Worm shape” etching pattern can be observed.

## 4. Rejuvenation Mechanism



Rejuvenator types influence the morphology of aged PMB

## 4. Rejuvenation

### Mechanism

Aging state of PMB

Rejuvenator Dosage



Rejuvenator Type

Addition of Fresh PMB



The higher rejuvenator content results in worm structure

## 4. Rejuvenation

### Mechanism

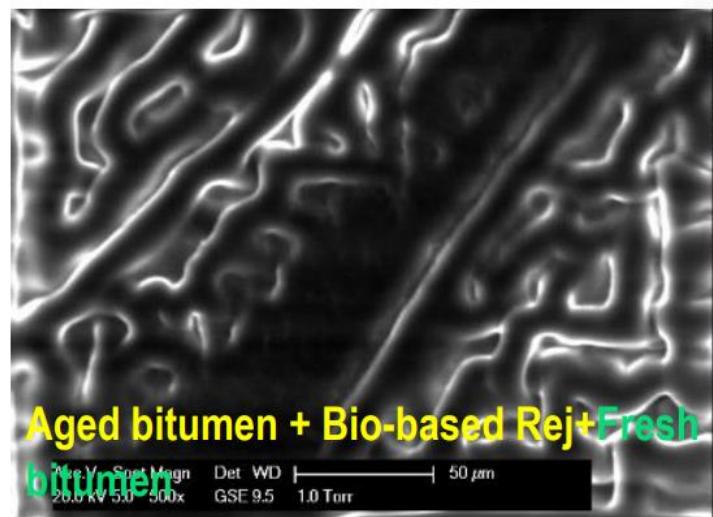
Aging state of PMB

Rejuvenator Dosage



Rejuvenator Type

Addition of Fresh PMB



## 4. Rejuvenation

### Mechanism

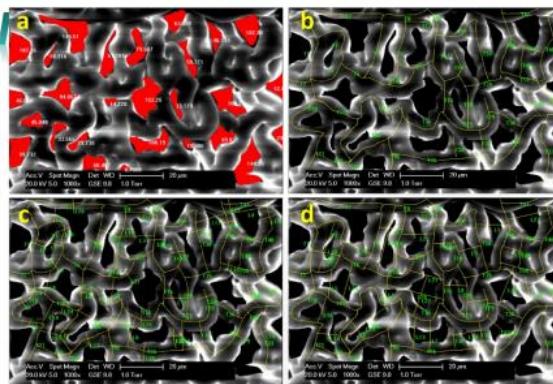


Image Pro Software to extract microstructure parameters:  
(a: area coverage; b: length; c: diameter; d: distance...)

Lin, Peng, X. Liu, P. Apostolidis, S. Erkens, Y. Zhang, and S. Ren. "ESEM Observation and Rheological Analysis of Rejuvenated SBS Modified Bitumen." *Materials & Design* 204 (June 1, 2021): 109639.

## Conclusions

- Recipe Optimization

- Recipe optimization should consider material selection, component ratio determination and **functional property balanced at different temperature conditions**.
- **SBS-based rejuvenator** is one of the most effective rejuvenators for the aged PMB binders.
- SBS based rejuvenator shows advantages in multi-cycles recycling capability.
- **Bio-based** rejuvenator shows advantage in reducing  $G^*$  at low temperature, but **drawbacks in aging resistance and viscoelastic property**.
- Aromatic based rejuvenator shows advantage in viscoelastic property, but drawback in other properties.
- EBA, TB, TB/SBS rejuvenators show advantage in high-temp property, but drawback in other properties.

# Conclusions

- Evaluation methods

- A series of **performance-based experimental methods** are selected for evaluation of rejuvenator effectiveness, dosage and multi-cycle recycling capability.
- Dosage can be determined with performance-based method instead of traditional methods.
- **MSCR and rejuvenation index evaluation** are sufficient to determine the rejuvenator dosage.
- **Multi-cycles recycling capability** evaluation is important for determination of effectiveness of rejuvenators.

# Conclusions

- Rejuvenation Mechanism

- **Fully understanding the rejuvenation mechanism is extremely important** for rejuvenator recipe optimization and effective evaluation methods development.
- **E-SEM is a useful tool** for rejuvenation mechanism study, as it can detect the morphology structures of the rejuvenated bitumen.
- Morphology of rejuvenated PMB is influenced not only by the aging degree, rejuvenator dosage/types, but also the addition of the fresh PMB.
- There is a close link between the chemical composition, rheological property and morphology structure in E-SEM.

Workshop: Changes in binder properties and the role of additives

**Thank you !**

