This document is the accepted manuscript version of the following article: Li, R., Leng, Z., Partl, M. N., & Raab, C. (2021). Characterization and modelling of creep and recovery behaviour of waterborne epoxy resin modified bitumen emulsion. Materials and Structures, 54(1), 8 (12 pp.). https://doi.org/10.1617/s11527-020-01594-6

Characterization and Modelling of Creep and Recovery Behaviour of Waterborne Epoxy Resin Modified Bitumen Emulsion

Rui Li^a, Zhen Leng^{a*}, Manfred N. Partl^b, Christiane Raab^b

^a Department of Civil and Environmental Engineering, The Hong Kong Polytechnic University, Hong Kong SAR, China

^b Laboratory of Road Engineering, Empa Swiss Federal Laboratories for Materials Science and Technology, Dübendorf, Switzerland

Abstract: Bitumen emulsion has been widely used as the tack coat and surface treatment material for asphalt pavement. A major advantage of this material is it does not require heating during construction. However, it faces the concern of low mechanical strength, especially at high service temperatures. To improve the mechanical strength of bitumen emulsion, various research efforts have been made to use waterborne epoxy resin as a modifier to produce waterborne epoxy modified bitumen emulsion (WEB). To better understand WEB as a paving material, this study aims to investigate the microstructure and evaluate the high-temperature performance and model the creep and recovery behaviour of WEB residues. First, a confocal laser scanning microscopy (CLSM) was employed to investigate the fluorescence microstructure of the WEB residues. The temperature sweep dynamic shear modulus tests were then conducted to characterize the viscoelastic properties of the WEB residues within the temperature range of 4 °C to 76 °C, and multiple stress creep recovery (MSCR) tests at 50 °C, 60 °C and 70 °C were conducted to evaluate their high temperature performance. Then, the complex modulus was fitted with Arrhenius model and the activation energy was calculated; and the creep and recovery test results were modelled with both the power law model and generalized Burgers model. Finally, the average percent recovery and non-recoverable compliance were determined. It was found that the rutting resistance of bitumen emulsion can be dramatically increased by incorporating waterborne epoxy resin, and the creep and recovery behaviour of bitumen emulsion can be better fitted by the generalized Burgers model in comparison with the power law model.

 $E\text{-}mail\ address:\ zhen.leng@polyu.edu.hk$

^{*} Corresponding author.

Keywords: Bitumen emulsion; Waterborne epoxy resin; Microstructure; Creep and recovery; Power

law model; Generalized Burgers model

1 Introduction

Asphalt pavement is major type of pavement worldwide due to its various advantages, such as driving

comfortability, low noise and ease of maintenance [1]. However, in the construction process of

conventional asphalt cement based hot-mix asphalt (HMA), volatile organic compounds (VOC) and

greenhouse gases (GHG) are produced, which adversely influence the environment [2]. On the other

hand, the water-based bitumen emulsion, which can be used at ambient temperature without heating,

produces much less hazardous emissions, leading to a greener binder material compared with asphalt

cement. However, bitumen emulsion faces the major concern of low mechanical strength at high

service temperatures [3].

To overcome this limitation, polymer latexes including styrene-butadiene-styrene (SBS) latex [4],

styrene-butadiene rubber (SBR) latex [5] and chloroprene rubber (CR) latex [6] have been applied to

modify bitumen emulsion. The advantage of polymer latexes is that they can be easily mixed with

bitumen emulsion through simple stirring in a plant or before application. Takamura [7] reported that

SBR latex was dispersed in the aqueous phase after its mixing with bitumen emulsion, and it formed a

continuous polymer film around the bitumen particles upon the curing of the emulsion. Such findings

are in consistent with those of Forbes, et al. [8]. The continuous polymer structure is deemed to be

responsible for the performance improvement of the residual binder. The incorporation of waterborne

epoxy resin into bitumen emulsion is a relatively new approach to modify bitumen emulsion, and the

relevant research is very limited. In one of the previous studies [9], waterborne epoxy resin and SBR

latex were mixed with bitumen emulsion simultaneously as modifiers. The results showed that both

the softening point and ductility of the bitumen emulsion residue were improved to some extent. More

recently, a waterborne epoxy resin was synthesized to modify bitumen emulsion, and the results

indicated that the high-temperature performance, bitumen-aggregate adhesion, resistance to moisture

damage, and resistance to fatigue were all improved substantially [10, 11].

2

As a viscoelastic material, bitumen displays time-dependent behavior, and the creep and recovery test has been commonly used to characterize this behaviour [12, 13]. When bitumen or bituminous binder is subjected to a constant load, deformation will occur and develop over time. The deformation can then partially recover upon the removal of the load [14]. The generalized Burgers model has been found to be a suitable model to approximate the creep and recovery behaviour of bitumious materials [15, 16]. The power law model, which requires less parameters, has also been applied for such purpose [17].

With the addition of waterborne epoxy resin, the viscoelastic behaviour of bitumen emulsion would change substantially after curing of epoxy resin. As a relatively new material, the viscoelastic performance of waterborne epoxy modified bitumen emulsion (WEB) has not been well understood yet. Thus, this study has been conducted aiming to investigate and model the viscoelastic behaviour of WEB residues, especially at high service temperatures.

2 Materials and methodology

2.1 Materials

Base bitumen with a penetration grade of 60/70, commonly used in Hong Kong, and the cationic type bitumen emulsifier INDULIN® W-5 supplied by Ingevity, a brown liquid mainly composed of amines and lignin, were used to prepare bitumen emulsion in this study. The basic properties of the base bitumen are shown in Table 1. To prepare bitumen emulsion, base bitumen was preheated to 150 °C. Meanwhile, by mixing emulsifier and deionized water at 60 °C, and with the addition of HCl to a pH value of 2.0-2.5, the so-called "soap water" was prepared. The amount of emulsifier was 3% by weight of bitumen as suggested by the supplier. The preheated bitumen and "soap water" were then emulsified in a colloid mill at a speed of 2,840 rpm to produce the bitumen emulsion with a solid content of 63.0% by weight.

The waterborne curing agent (WCA) was used to emulsify epoxy resin for producing the epoxy emulsion following the procedure described by Leng et al. [10]. It is interesting to note that the WCA can also act as an emulsifier for epoxy resin. The WCA was mixed with epoxy resin in a three-necked

flask to produce the epoxy emulsion with a solid content of 60% by weight. Based on the stoichiometric calculation, the weight ratio of epoxy resin and WCA was determined to be 4:3. Deionized water was dropped slowly using a separating funnel during the stirring of the two parts of epoxy emulsion. This process was continued by stirring at a speed of 700-800 rpm for 5 min at ambient temperature.

The bitumen emulsion and epoxy resin emulsion were mixed and stirred at a speed of 200-300 rpm for about 10 min to prepare a homogeneous WEB (Fig. 1). Fig. 2 represents a schematic structure of the WEB, where the bitumen particles are stabilized by cationic emulsifier, while the waterborne epoxy emulsion is distributed evenly in the water continuous phase. Epoxy resin emulsion were added into bitumen emulsion in three different percentages: 1%, 3% and 5%, and the corresponding WEBs were denoted as WEB-1, WEB-3 and WEB-5, respectively, in this study. All the percentages were based on the solid weight of emulsions. The bitumen emulsion without epoxy resin was also evaluated as a control material and denoted as WEB-0. A thin layer of WEB emulsion was poured on a silicone paper and maintained at ambient room condition for 3 days, followed by 12 h of oven heating at 60 °C. It has been verified in a previous study [10] that the epoxy resin will be completely cured after this procedure.

Table 1 Basic properties of base bitumen

Penetration (25 °C, 0.1 mm)	Softening point (°C)	Viscosity at 135 °C (mPa·s)		
64.5	50	477.5		

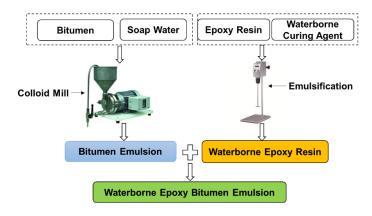


Fig. 1 Procedure of waterborne epoxy bitumen emulsion preparation

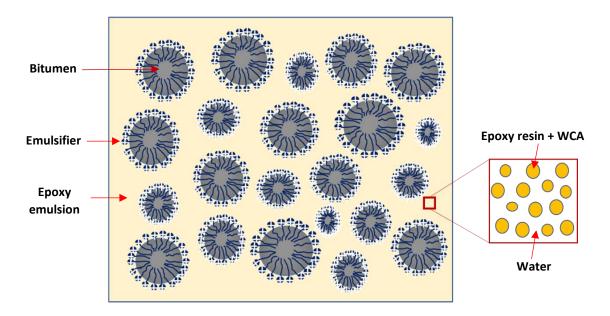


Fig. 2 Schematic structure of waterborne epoxy bitumen emulsion

2.2 Research program

The microstructure of the bitumen emulsion residues were first investigated with a Leica TCS SPE Confocal Microscope. The images were recorded in fluorescent mode using a laser with a wavelength of 488 nm (blue light). The emulsion residue was obtained by placing a drop of emulsion on a microscope slide and dried at ambient temperature for 3 h, the cover glass was then placed on the specimen, and finally the specimen was put into a forced air oven at 60 °C for 4 h to fully evaporate the water. Such preparation process of the specimen resembles that of the field application of bitumen emulsion especially for the pavement surface treating at the hot season, and thus can reflect the true microstructure of the bitumen emulsion residue.

Rutting is one of the main distresses of asphalt pavements affecting pavement service life, driving comfortability and safety. To characterize the rutting potential of bituminous binders, the Superpave rutting parameter has been developed and used in the US binder grading system. However, many studies have indicated that this parameter is not always adequate in representing the rutting potential of bituminous binders, especially the polymer modified binders [18-21]. This is because that polymer modified binders possess substantial delayed elastic response [20]. Correspondingly, the MSCR test has been proposed as an alternative approach to characterize the rutting potential of bitumen and polymer modified binders [22, 23]. This test is now listed in the standard specification of AASHTO

MP 19. The non-recoverable creep compliance (J_{nr}) derived from this test has been reported to correlate well with the performance of ordinary asphalt pavement mixtures [24-26].

To characterize the viscoelastic performance of WEB residues, dynamic shear rheometer (DSR) tests were conducted on the cured emulsion residues. Temperature sweep from 4 °C to 94 °C with 6 °C temperature increments was first applied in strain-controlled sinusoidal mode at a testing frequency of 10 rad/s within the linear viscoelastic region [27]. MSCR tests [28] were then conducted at three different temperatures: 50 °C, 60 °C and 70 °C. Two shear stress levels: 0.1 kPa and 3.2 kPa, were applied. Two viscoelastic models, namely the power law model and the generalized Burgers model, were applied to model the creep-recovery behaviour at 0.1 kPa. The average percent recovery and non-recoverable compliance were then computed to determine the rutting potential of the WEB residues.

2.3 Creep and compliance modelling

In the MSCR test, the lower stress of 0.1 kPa is adopted to evaluate the linear viscoelastic performance of bituminous binders, while the higher stress of 3.2 kPa is applied to characterize the nonlinear viscoelastic response of most bituminous binders [29]. In this study, the binders' responses at the lower stress of 0.1 kPa in the linear viscoelastic range were selected and modelled using the power low model and the generalized Burgers model.

2.3.1 Power law model

The power law model was considered because it has fewer material coefficients than the Burgers model. In this model, the strain response $\varepsilon(t)$ at time t can be expressed as:

$$\varepsilon(t) = (k + at^b) * \sigma^c \tag{1}$$

where k is the instant elastic strain response, a and b are the shape parameters, and c is a coefficient associated with the stress nonlinearity of the material. Based on the Boltzmann superposition principle, the strain response at the recovery phase is given as

$$\varepsilon(t) = \left(a(t^b - (t - t_0)^b\right) * \sigma^c \tag{2}$$

where t_0 is the time of the creep phase.

2.3.2 Generalized Burgers model

The generalized Burgers model, which consists of one Maxwell model and a number of Kelvin-Voigt model in series, has been widely used in modelling the viscoelastic behaviour of bituminous binders [30, 31]. The creep strain response $\varepsilon(t)$ is

$$\varepsilon(t) = \sigma D_0 + \sum_{i=1}^n \sigma D_i \left(1 - e^{-t/\tau_i} \right) + \frac{\sigma}{\mu_0} t \tag{3}$$

The strain in the recovery phase is given as

$$\varepsilon(t) = \sum_{i=1}^{n} \sigma D_i e^{-t/\tau_i} \left(e^{t_0/\tau_i} - 1 \right) + \frac{\sigma}{\mu_0} t_0 \tag{4}$$

$$\varepsilon(t) = \sigma \cdot \left(\frac{t_0}{\mu_0} + \sum_{i=1}^n D_i e^{-t/\tau_i} \left(e^{t_0/\tau_i} - 1\right)\right)$$

where σ is the applied stress, kPa, t is the current time, D_0 is the creep compliance of the standalone spring, D_i is the creep compliance of the i^{th} Kevin series, τ_i is the retardation time of the i^{th} Kevin series, n is the number of Kevin series, t_0 is the time of creep phase, and μ_0 is the zero shear viscosity [21, 32].

The least-squre curve fitting method in MATLAB® program was applied in both models to fit all creep and recovery data.

3 Results and discussions

3.1 Microstructure of WEB residues

Fig.3 depicts the fluorescence images of the cured WEB residues. The dark phase corresponds to bitumen, and the yellow phase corresponds to epoxy resin. It can be seen that with the increase of the waterborne epoxy resin dosage, the area of the yellow phase increased. It is also interesting to notice that the cured epoxy resin forms an inter-connected polymer (or honeycomb-like) structure when the dosage of the waterborne epoxy resin reached 3wt%, and the bitumen filled the honeycomb holes.

Such honeycomb-like structure is similar with that found by Takamura [33] on the residue of SBR latex modified bitumen emulsion. The waterborne epoxy resin emulsion was dispersed in the aqueous phase after mixing with bitumen emulsion. As water evaporates and the bitumen droplets become closer, the epoxy resin particles are squeezed to form a thin layer around the bitumen particles. After the curing process of the epoxy resin, a continuous epoxy film will be formed surrounding the bitumen particles. This change of the structure may significantly affect the viscoelastic properties of the bitumen binder, which will be further discussed in the following parts of this paper.

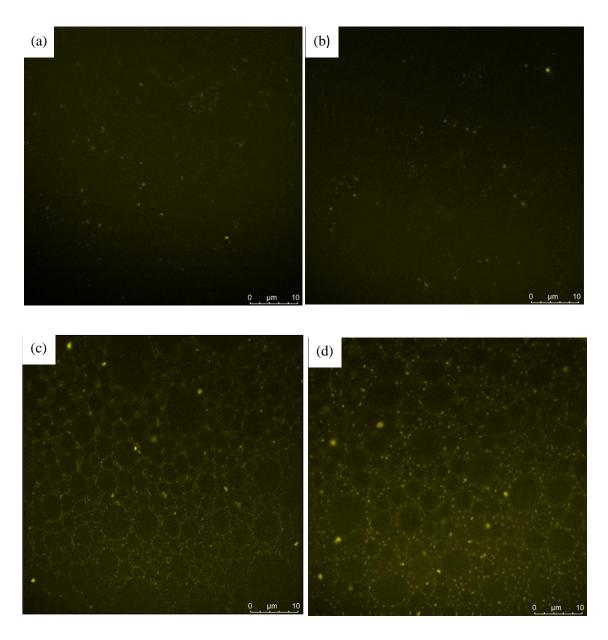


Fig. 3 CLSM images of WEB residues: (a) WEB-0; (b) WEB-1; (c) WEB-3; (d) WEB-5 (scale bar: 10 µm)

3.2 Temperature dependence of complex modulus

The viscoelastic performance of bitumen is highly temperature dependent. It has been reported that the Arrhenius model captures the temperature-dependence of bituminous materials well [34]. The so-called activation energy E_a can be used as an important parameter to evaluate the temperature susceptibility of bitumen. Larger values of E_a represent higher temperature sensitivities, while lower values of E_a indicate lower temperature sensitivities [35, 36]. The Arrhenius type of equation was developed and adopted to characterize the temperature dependence of bitumen viscosity [36, 37]. E_a for flow generally decreases with the increase of polymer content in bitumen binders [34, 35]. Later, the Arrhenius equation was extended to describe the dynamic complex modulus as a function of temperature, which was found to have good agreement with experiment results [38, 39]. The Arrhenius equation for the complex modulus is given as

$$|G^*| = Ae^{\frac{E_a}{RT}} \tag{5}$$

It can be transferred to the following formula by taking logarithm on both sides,

$$\ln|G^*| = \frac{E_a}{RT} + \ln A \tag{6}$$

where A is a pre-exponential parameter, G^* is the complex modulus, E_a is the activation energy, $R = 8.314 J * K^{-1} * mol^{-1}$ is the universal gas constant, and T is the temperature in Kelvin.

As Fig. 4 demonstrates, the complex moduli of all bitumen emulsion residues can be well fitted using the Arrhenius model with high R^2 values of 0.988 and 0.998. The values of activation energy E_a obtained from the modelling are presented in Table 2. It can be seen that WEB-0 has the highest E_a value of 125.8 kJ/mol. With the increase of waterborne epoxy content, the values of E_a decreased, indicating that the temperature susceptibility of emulsion residues was reduced by incorporating waterborne epoxy resin. The range of activation energy is similar to the results reported by previous studies [36, 38].

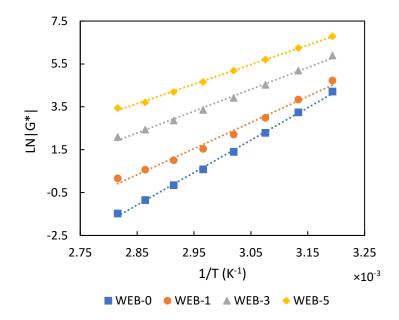


Fig. 4 Arrhenius plot of complex modulus of WEB residues

Table 2 Activation energy based on the Arrhenius model

WEB-residues	Ea (kJ/mol)	\mathbb{R}^2
WEB-0	125.8	0.998
WEB-1	101.2	0.988
WEB-3	84.5	0.995
WEB-5	75.7	0.998

3.3 Creep and recovery modelling

3.2.1 Power law model

The power law model was first applied to fit the creep and recovery test results at 0.1 kPa in the present study. It was found by trial that the power parameter c is close to 1, indicating linearity of stress dependence. In addition, the instant elastic strain response k is very close to 0 (mostly in the scale of 1e-7). Thus, the power law model was reduced to

$$\varepsilon(t) = \sigma * at^b \tag{5}$$

Fig. 5 shows one cycle of creep and recovery at 0.1 kPa for all the WEB residues, and Table 3 presents the fitting parameters of the power law model. The solid lines in Fig. 5 represent the fitted curves using the power law model. It can be seen that the data fitting for WEB-0 is excellent, while

the fitting curve deviates from the measured data for WEB-1, WEB-3 and WEB-5. The elliptical circles in these figures highlight the areas where the fitting curve deviates from the measured data.

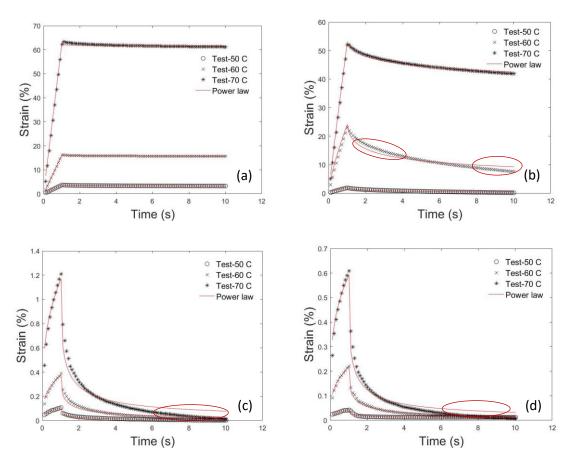


Fig. 5 Power law fitting ((a) WEB-0, (b) WEB-1, (c) WEB-3, (d) WEB-5)

Table 3 Fitting parameters of the power law model

Binder	Temperature (°C)	a	b	\mathbb{R}^2
	50	36.44	0.97	0.9996
WEB-0	60	159.75	1.00	0.9993
	70	620.69	1.00	0.9959
WEB-1	50	20.32	0.61	0.8240
	60	239.49	0.72	0.9149
	70	525.10	0.94	0.9936
WEB-3	50	1.05	0.33	0.9807
	60	3.73	0.35	0.9548
	70	11.62	0.31	0.9640
WEB-5	50	0.27	0.73	0.9068
	60	2.16	0.33	0.9898
	70	5.89	0.28	0.9765

3.2.2 Generalized Burgers model

Apart from the power law model, the generalized Burgers model was also applied as a possible alternative for modeling the data. It was found that the original Burgers four element model, which contains one Kelvin model, was inadequate to well fit the data, especially for the recovery phase at higher temperature. When adding one more Kelvin model in series, it can be seen from Fig. 6 that excellent fitting can be achieved. The fitting parameters are presented in Table 4, which shows that the R² values are larger than 0.99 for all cases.

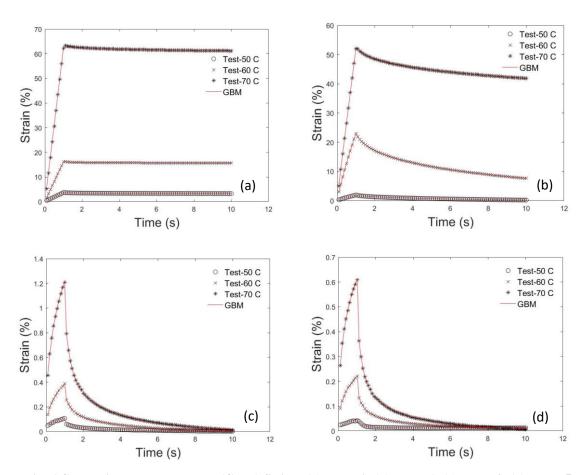


Fig. 6 Generalized Burgers model (GBM) fitting: (a) WEB-0, (b) WEB-1, (c) WEB-3, (d) WEB-5

Table 4 Fitting parameters for the generalized Burgers model

Binder	Temperature (°C)	D_0	μ_0	D_1	$ au_1$	D_2	$ au_2$	\mathbb{R}^2
WEB-0	50	0.75	0.034	79.98	1.00	13.81	256.46	0.9999
	60	0.01	0.006	10.00	1.79	3.16	798.16	0.9983
	70	0.01	0.002	999.96	78.93	28.61	29.60	0.9925
WEB-1	50	1.82	0.667	33.00	33.00	3.61	3.61	0.9998
	60	3.80	0.022	53.15	900.10	0.63	6.01	0.9993
	70	0.01	0.003	834.38	49.49	7.86	0.88	0.9969
WEB-3	50	0.01	21.78	3.40	0.01	0.39	7.81E-07	0.9997

	60	0.02	6.10	2.80	0.05	0.33	2.23E-14	0.9998
	70	0.07	2.47	2.65	0.18	0.33	1.67E-03	0.9997
	50	2.28E-03	174.96	0.27	3.50E-03	0.78	3.88E-03	0.9999
WEB-5	60	0.02	15.84	2.54	0.03	0.34	2.95E-03	0.9998
	70	0.05	6.17	2.35	0.09	0.30	1.82E-03	0.9998

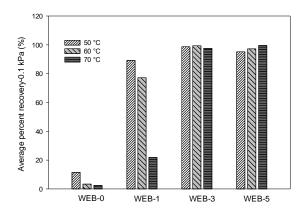
In the generalized Burgers model, the viscosity parameter μ_0 of the single dashpot represents the zero shear viscosity (ZSV). ZSV has been taken as an effective indicator for permanent deformation of bituminous binders by many researchers [40-42]. Larger values of ZSV indicate stronger resistance to permanent deformation. Table 4 shows that the values of ZSV generally increased with the increase of epoxy resin content and decreased with the increasing temperature. Similar results were reported in other studies [25, 32]. Such phenomenon is consistent with the microstructure observed in Fig. 3, as the inter-connected polymer structure can increase the residue's viscosity substantially. In addition, the formation of the inter-connected polymer structure also increased the elasticity of the emulsion residue, thus the fitted parameters of creep compliance (D_1 , D_2) and retardation time (T_1 , T_2) generally decreased with increasing waterborne epoxy resin dosages.

Comparing the fitting results by the generalized Burgers model and the power law model, the latter model has only two parameters, which is simpler. However, the generalized Burgers model was more accurate for the creep and recovery behaviour modelling. For example, the R^2 for WEB-5 at 50 °C by the power law model was around 0.9, while it was very close to 1 by the generalized Burgers model. It should also be noted that the instant elastic parameter k was almost 0 for the power law model, while the instant elastic parameter D_0 in the generalized Burgers model was not. The mechanism behind such observation should be further studied in the future. In addition, it can be found that the parameter μ_0 from the generalized Burgers model was closely related to bitumen viscosity, and was consistent with the respective material's performance, i.e., μ_0 increased with larger dosages of waterborne epoxy resin and decreased with higher temperatures. However, most other parameters did not show a particular trend, and may not be relate to materials actual properties.

3.4 MSCR analysis

Fig. 7 shows the average percent recovery (R) calculated at the stress levels of 0.1 kPa and 3.2 kPa. The data were the average of 10 cycles of percent recovery, and the standard deviation was within 5% of the mean values. It can be seen that the percent recovery generally increased with the increase of the waterborne epoxy resin content. In addition, the percent recovery showed a decreasing trend with the increasing temperature, except for those of WEB-3 and WEB-5 at 0.1 kPa. In fact, at the stress level of 0.1 kPa, the percent recovery for WEB-0 was less than 15% even at 50 °C, while those of WEB-3 and WEB-5 reached more than 90% at the same temperature. The recovery difference between different temperatures indicates temperature susceptibilities. It is apparent that the recovery difference was much lower when larger amount of waterborne epoxy resin was used. The difference can be explained by the microstructure of the WEB residues. As shown in Fig. 3, the inter-connected epoxy polymer structure was formed when the waterborne epoxy resin dosage was increased to 3wt%, while such connected polymer structure was not observed in WEB-1. This connected polymer structure increased the residue's recovery ability, and decreased the temperature sensitivity.

Moreover, it was found in Fig. 7 that the strain recovery at 3.2 kPa decreased compared with that at 0.1 kPa. The larger difference of recovery between different stresses means higher stress sensitivity. However, the percent recoveries for WEB-3 and WEB-5 did not change much at the two stress levels, while those of WEB-0 and WEB-1 decreased significantly. Thus, it can be inferred that larger quantities of waterborne epoxy could lead to lower stress sensitivity of the emulsion residues because of the formation of the inter-connected epoxy polymer structure.



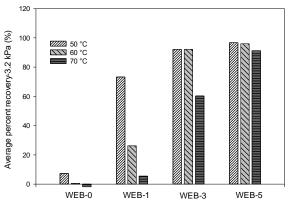


Fig. 7 Average percent recovery of MSCR test (left-0.1 kPa, right-3.2 kPa)

The non-recoverable creep compliance (J_{nr}) results are shown in Fig. 8. It is clear that J_{nr} decreased dramatically with the increasing waterborne epoxy resin content. The most distinct decrease of J_{nr} was observed in WEB-3 and WEB-5 at all test temperatures and stress levels. In addition, J_{nr} generally increased with the increase of temperature and stress. The non-recoverable compliance J_{nr} has been used as an important indicator for the rutting sensitivity. Lower J_{nr} indicates stronger resistance to rutting. It is suggested that a value of J_{nr} below 0.5 kPa⁻¹ at a stress level of 3.2 kPa represent good resistance to permanent deformation [24]. A closer look at Fig. 8 reveals that the values of J_{nr} of WEB-3 and WEB-5 were both less than 0.02 kPa⁻¹ even at the stress of 3.2 kPa, substantially lower than that of WEB-0 and WEB-1. Therefore, it can be concluded that waterborne epoxy resin remarkably improved the rutting resistance of bitumen emulsion residues, and a minimum of 3wt% waterborne epoxy resin is a reasonable requirement for obtaining good rutting resistance. The big difference between WEB-3&WEB-5 and that of WEB-0&WEB-1 is also related with the presence of the inter-connected polymer structure. For WEB-3 and WEB-5, the cured epoxy resin formed a continuous polymer-rich film, and the bitumen particles are capsuled in the film, which turned the thermal-plastic bitumen into a thermal-setting composite material. Thus, the resistance to permanent deformation was significantly improved.

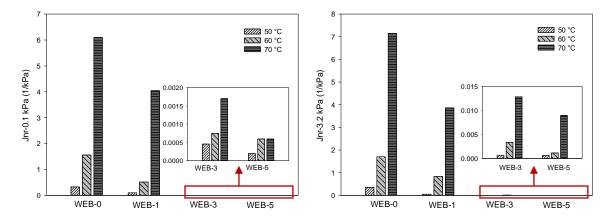


Fig. 8 Non-recoverable creep compliance J_{nr} of MSCR test (left-0.1 kPa, right-3.2 kPa, a close up was shown for WEB-3 and WEB 5)

4 Findings and conclusions

In this study, waterborne epoxy resin was used as a strength enhancing modifier for bitumen emulsion. The microstructures of the waterborne epoxy bitumen (WEB) residues were first evaluated by the fluorescence microscopy. The rheological properties of the emulsion residues were then characterized through temperature sweep of the complex modulus and MSCR tests, and the MSCR at 0.1 kPa was modelled through both the power law model and the generalized Burgers model. Based on the test and modeling results, the following major findings have been obtained:

- The fluorescence microscopy reveals that the cured waterborne epoxy resin forms an interconnected polymer film around the bitumen particles when the epoxy dosages reached 3wt%.
- The Arrhenius model described the complex modulus of waterborne epoxy bitumen (WEB) residues as a function of temperature very well, and the activation energy can be used to evaluate the temperature sensitivity of binders.
- Both the power law model and the generalized Burgers model were able to fit the creep
 and recovery data of WEB residues in a reasonable way. The power law model used less
 parameters, while the generalized Burgers model can fit the creep-recovery data more
 accurately.
- With the increase of waterborne epoxy resin concentration, the percent recovery of WEB significantly increased, and the non-recoverable compliance decreased greatly, suggesting that waterborne epoxy resin improved the resistance to rutting of bitumen emulsion residues remarkably. For example, compared with WEB-0, the percent recovery of WEB-5 increased to more than 90% at all testing temperatures and stresses, while its non-recoverable compliance decreased to less than 0.02 kPa⁻¹.

Overall, the findings from this research suggested that the high-temperature performance of bitumen emulsion residues can be significantly improved by adding waterborne epoxy resin. Waterborne epoxy resin modified bitumen emulsion can be potentially applied as the material for tack coat,

pavement surface treatment, cold recycling of reclaimed asphalt pavement (RAP), and cold mix asphalt. Further studies should be conducted to explore other properties of WEB, such as aging resistance, adhesion between emulsion residue and aggregates, and low temperature and fatigue cracking resistance.

Acknowledgement

The authors would like to acknowledge the sponsorship of the Hong Kong Innovation and Technology Commission through the Guangdong-Hong Kong Technology Cooperation Funding Scheme (TCFS) project GHP/116/18GD, as well as the PhD research student attachment program of the Hong Kong Polytechnic University.

References

- [1] M. García-Morales, P. Partal, F. J. Navarro, and C. Gallegos, Effect of waste polymer addition on the rheology of modified bitumen, *Fuel*, vol. 85, no. 7, pp. 936-943, 2006.
- [2] S. D. Capitão, L. G. Picado-Santos, and F. Martinho, Pavement engineering materials: Review on the use of warm-mix asphalt, *Construction and Building Materials*, vol. 36, pp. 1016-1024, 2012/11/01/ 2012.
- [3] B. Gómez-Meijide and I. Pérez, A proposed methodology for the global study of the mechanical properties of cold asphalt mixtures, *Materials & Design*, vol. 57, pp. 520-527, 2014/05/01/ 2014.
- [4] H. M. Cai, T. Wang, J. Y. Zhang, and Y. Z. Zhang, Preparation of an SBS Latex–Modified Bitumen Emulsion and Performance Assessment, *Petroleum Science and Technology*, vol. 28, no. 10, pp. 987-996, 2010/05/17 2010.
- [5] A. Khadivar and A. Kavussi, Rheological characteristics of SBR and NR polymer modified bitumen emulsions at average pavement temperatures, *Construction and Building Materials*, vol. 47, pp. 1099-1105, 2013/10/01/ 2013.
- [6] J. Jiang, F. Ni, J. Zheng, Y. Han, and X. Zhao, Improving the high-temperature performance of cold recycled mixtures by polymer-modified asphalt emulsion, *International Journal of Pavement Engineering*, vol. 21, no. 1, pp. 41-48, 2020/01/02 2020.
- [7] K. Takamura, "Microsurfacing with SBR latex modified asphalt emulsion," Charlotte, NC, BASF, 2002.
- [8] A. Forbes, R. G. Haverkamp, T. Robertson, J. Bryant, and S. Bearsley, Studies of the microstructure of polymer-modified bitumen emulsions using confocal laser scanning microscopy, *Journal of Microscopy*, vol. 204, no. 3, pp. 252-257, 2001.
- [9] R. Zhang, Y. He, and Z. Ao, An asphalt emulsion modified by compound of epoxy resin and styrene-butadiene rubber emulsion, *International Journal of Mathematical Models and Methods in Applied Sciense*, no. 4, 2007.
- [10] R. Li, Z. Leng, Y. Zhang, and X. Ma, Preparation and characterization of waterborne epoxy modified bitumen emulsion as a potential high-performance cold binder, *Journal of Cleaner Production*, 2019.

- [11] C. Hu, J. Zhao, Z. Leng, M. N. Partl, and R. Li, Laboratory evaluation of waterborne epoxy bitumen emulsion for pavement preventative maintenance application, *Construction and Building Materials*, vol. 197, pp. 220-227, 2019/02/10/ 2019.
- [12] P. Shirodkar, Y. Mehta, A. Nolan, K. Dahm, R. Dusseau, and L. McCarthy, Characterization of creep and recovery curve of polymer modified binder, *Construction and Building Materials*, vol. 34, pp. 504-511, 2012.
- [13] M. Elnasri, G. Airey, and N. Thom, "Experimental investigation of bitumen and mastics under shear creep and creep-recovery testing," in *Airfield and Highway Pavement 2013: Sustainable and Efficient Pavements*, 2013, pp. 921-932.
- [14] F. Bai, X. Yang, and G. Zeng, Creep and recovery behavior characterization of asphalt mixture in compression, *Construction and Building Materials*, vol. 54, pp. 504-511, 2014/03/15/ 2014.
- [15] N. Saboo and A. Mudgal, Modelling creep and recovery response of asphalt binders using generalized burgers model, *Petroleum Science and Technology*, vol. 36, no. 20, pp. 1627-1634, 2018/10/18 2018.
- [16] N. Saboo, A. Mudgal, and A. Singh, Using Frequency Sweep Test to Predict Creep and Recovery Response of Asphalt Binders, *Journal of Materials in Civil Engineering*, vol. 31, no. 6, p. 04019081, 2019.
- [17] N. Saboo and P. Kumar, A study on creep and recovery behavior of asphalt binders, *Construction and Building Materials*, vol. 96, pp. 632-640, 2015.
- [18] K. D. Stuart, W. S. Mogawer, and P. Romero, "Evaluation of the Superpave asphalt binder specification for high-temperature pavement performance," in *Association of Asphalt Paving Technologists Proc*, 2000, vol. 69.
- [19] H. U. Bahia, D. Hanson, M. Zeng, H. Zhai, M. Khatri, and R. Anderson, *Characterization of modified asphalt binders in superpave mix design* (no. Project 9-10 FY'96). 2001.
- [20] J. D'Angelo and R. Dongre, "Superpave binder specifications and their performance relationship to modified binders," in *Proceedings of the Forty-Seventh Annual Conference of the Canadian Technical Asphalt Association (CTAA): Calgary, Alberta*, 2002.
- [21] S. Biro, T. Gandhi, and S. Amirkhanian, Determination of zero shear viscosity of warm asphalt binders, *Construction and Building Materials*, vol. 23, no. 5, pp. 2080-2086, 2009/05/01/2009.
- [22] J. D'Angelo, Current status of Superpave binder specification, *Road Materials and Pavement Design*, vol. 10, no. sup1, pp. 13-24, 2009.
- [23] J. A. D'Angelo, The Relationship of the MSCR Test to Rutting, *Road Materials and Pavement Design*, vol. 10, no. sup1, pp. 61-80, 2009/01/01 2009.
- [24] E. DuBois, D. Y. Mehta, and A. Nolan, Correlation between multiple stress creep recovery (MSCR) results and polymer modification of binder, *Construction and Building Materials*, vol. 65, pp. 184-190, 2014/08/29/ 2014.
- [25] O.-V. Laukkanen, H. Soenen, T. Pellinen, S. Heyrman, and G. Lemoine, Creep-recovery behavior of bituminous binders and its relation to asphalt mixture rutting, *Materials and Structures*, journal article vol. 48, no. 12, pp. 4039-4053, December 01 2015.
- [26] J. Zhang, L. F. Walubita, A. N. M. Faruk, P. Karki, and G. S. Simate, Use of the MSCR test to characterize the asphalt binder properties relative to HMA rutting performance A laboratory study, *Construction and Building Materials*, vol. 94, pp. 218-227, 2015/09/30/2015.
- [27] Determining the rheological properties of asphalt binder using a dynamic shear rheometer (DSR), 2013.
- [28] Standard method of test for multiple stress creep recovery (MSCR) test of asphalt binder using a dynamic shear rheometer (DSR), 2013.
- [29] A. Behnood and J. Olek, Rheological properties of asphalt binders modified with styrene-butadiene-styrene (SBS), ground tire rubber (GTR), or polyphosphoric acid (PPA), *Construction and Building Materials*, vol. 151, pp. 464-478, 2017.

- [30] M. F. Woldekidan, "Response modelling of bitumen, bituminous mastic and mortar," Road and Railway Engineering, Delft University of Technology, 2011.
- [31] C. Celauro, C. Fecarotti, A. Pirrotta, and A. C. Collop, Experimental validation of a fractional model for creep/recovery testing of asphalt mixtures, *Construction and Building Materials*, vol. 36, pp. 458-466, 2012/11/01/ 2012.
- [32] F. Giuliani, F. Merusi, and I. Antunes, "Creep flow behavior of asphalt rubber binder—The zero shear viscosity analysis," in *Proceedings of the Asphalt rubber*, 2006.
- [33] K. Takamura, "Pavement Preservation Using the SBR Latex Modified Asphalt Emulsion, BASF Corporation," in *International Latex Conference, Charlotte, NC*, 2005.
- [34] M. García-Morales *et al.*, Viscous properties and microstructure of recycled eva modified bitumen, *Fuel*, vol. 83, no. 1, pp. 31-38, 2004/01/01/ 2004.
- [35] D. Salomon and H. Zhai, Ranking asphalt binders by activation energy for flow, *Journal of Applied Asphalt Binder Technology*, vol. 2, no. 2, pp. 52-60, 2002.
- [36] S. W. Haider, M. W. Mirza, A. K. Thottempudi, J. Bari, and G. Y. Baladi, "Characterizing temperature susceptibility of asphalt binders using activation energy for flow," in *Transportation and Development Institute Congress 2011: Integrated Transportation and Development for a Better Tomorrow*, 2011, pp. 493-503.
- [37] I. A. Hussein, M. H. Iqbal, and H. I. Al-Abdul-Wahhab, Influence of Mw of LDPE and vinyl acetate content of EVA on the rheology of polymer modified asphalt, *Rheologica Acta*, journal article vol. 45, no. 1, pp. 92-104, September 01 2005.
- [38] G. Airey, M. Mohammed, A. C. Collop, C. J. Hayes, and T. Parry, Linear Viscoelastic Behaviour of Polyacrylate Binders and Bitumen Blends, *Road Materials and Pavement Design*, vol. 9, no. sup1, pp. 13-35, 2008/01/01 2008.
- [39] S. A. Narayan, D. N. Little, and K. Rajagopal, Incorporating Disparity in Temperature Sensitivity of Asphalt Binders into High-Temperature Specifications, *Journal of Materials in Civil Engineering*, vol. 31, no. 1, p. 04018343, 2018.
- [40] M. Phillips and C. Robertus, "Binder rheology and asphaltic pavement permanent deformation; the zero-shear-viscosity," in *EURASPHALT & EUROBITUME CONGRESS, STRASBOURG, 7-10 MAY 1996. VOLUME 3. PAPER E&E. 5.134*, 1996.
- [41] C. Binard, D. Anderson, L. Lapalu, and J. Planche, "Zero shear viscosity of modified and unmodified binders," in *PROCEEDINGS OF THE 3RD EURASPHALT AND EUROBITUME CONGRESS HELD VIENNA, MAY 2004*, 2004, vol. 2.
- [42] F. Morea, J. O. Agnusdei, and R. Zerbino, Comparison of methods for measuring zero shear viscosity in asphalts, *Materials and Structures*, journal article vol. 43, no. 4, pp. 499-507, May 01 2010.