Study on rheological properties of EVA modified asphalt based on fractional derivative theory

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Abstract: Based on the fractional derivative theory, the relation between fractional derivative operator and power function empirical creep equation has been established. And the physical meaning of the power function parameters has been clarified. Ethylene-vinyl acetate (EVA) has been used as additives to prepare modified asphalt with content of EVA modifier varying from 0% to 9% in increments of 3% by weight of asphalt. Dynamic frequency sweep tests have been conducted on EVA modified asphalt using dynamic shear rheometer at 30ºC, 40ºC, 50ºC, and 60ºC. The complex shear modulus and the rutting factor of the asphalt modified by EVA increase with the increase of EVA content and decrease with the increase of temperature. The results indicate that the EVA modified asphalt reduced thermal sensitivity and increased resistance to permanent at high temperatures. Also, the power function parameters of EVA modified asphalt have been studied and analysed based on fractional derivative theory.

Keywords: fractional derivative; power function empirical creep equation; EVA modified asphalt; dynamic shear rheological tests; frequency sweep; high temperature stability.

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1 Introduction

Polymer modifiers are used to improve the high-temperature stability of bituminous binders. There are three kinds of polymer modified asphalt: thermoplastic elastomeric modified asphalt, rubber modified asphalt and resin modified asphalt. Among them, resin modified asphalt mainly includes ethylene-vinyl acetate (EVA) modified asphalt, polyethylene (PE) modified asphalt and other materials. Polymer modified bitumen using a 60/70-penetration grade bitumen and recycled EVA/LDPE was prepared under different processing conditions (García-Morales et al., 2006). Measurements of the evolution of viscosity with time were carried out with rheomixer. Low agitation speeds and a processing temperature of around 180°C should be chosen for bitumen modification with the polymer used. This finding provides some technical guidance for the preparation of polymer modified asphalt.

Commonly used polymers are EVA plastomer and styrene-butadiene-styrene (SBS) elastomer. EVA can lower temperature susceptibility and rutting. SBS-D polymer modification can increased the complex modulus and elastic response of bitumens, particularly at high temperatures and low frequencies (Airey, 2002, 2003). So, modified bitumens were prepared by mixing two types of styrene-butadiene-styrene (SBS-D and SBS-M) and EVA with B 160/220 base bitumen 3% by weight. Then, Dynamic Shear Rheometer (DSR) test were conducted at 40ºC, 50ºC, 60ºC and 70ºC and at ten different frequencies. The results showed that the shear complex modulus (G*) of unaged, short- and long-term aged binders reduced and the phase angle (δ) of those increased. The G*/sinδ values increased with the addition of modifiers to the base bitumen (Alatas et al., 2014). Fan et al. (2017) also researched the effect of VA content on rheology, compatibility and conventional properties of EVA modified asphalt. The study was carried out by using dynamic mechanical analysis (DMA) and fluorescence microscopy.

There are thousands of metric tons of waste-polyethylene (WPE) producing every year, but their disposal is particularly difficult. The incorporation of waste polymers to bitumen as a modifier is a method that may relieve this problem. Therefore, the rheological properties of waste polyethylene modified asphalt were studied and the results showed that WPE could improve the rheological properties of asphalt pavement but it was often beleaguered with other concerns such as storage stability and phase separation (Kumar and Anand, 2018; Hu et al., 2015). However, Yu et al. (2015) presented a performance study of asphalt modified using waste packaging polyethylene (WPE) and organic montmorillonite (OMT). The results showed that excellent high temperature rheological properties of modified asphalt did not reduced and the storage stability of WPE was improved for the addition of OMT. Moreover, Qiu Sheng and Gang (2015) studied the effect of adding PE and the results showed that asphalt softening point, the complex modulus and rutting factor improved significantly. The static modulus and dynamic stability of asphalt mixture adding PE were improved significantly.

Bitumen is a viscoelastic material that exhibits both elastic and viscous components of response and displays both a temperature and time dependent relationship between applied stresses and resultant strains. So the paper aims to use a DSR and fractional derivative theory to provide a more reasonable and accurate evaluation of the rheological properties of EVA modified asphalt. The relation between fractional-derivative operator and power function empirical creep equation will be established and the physical significance of the power function parameters were further clarified based on fractional derivative theory. Then, the parameters of the creep equation of power function will be determined by the dynamic frequency sweep test results of EVA modified asphalt.

2 Fractional derivative theory

2.1 The basic theory

The viscoelastic constitutive model of fractional derivative essentially reflects the memory effect. It requires fewer model parameters to provide better constitutive description in a wider time or frequency domain (Shimizu and Zhang, 1999; Zhang et al., 2002). Fractional derivative differential operator has a variety of definition forms. In basic mathematics and engineering application research, Riemann-Liouville definition, Caputo definition and Grünwald-letnikov definition (Gemant, 1938; Gaputo, 1966; Bagley and Torvik, 1983, 1986) of fractional derivative are most commonly used. Among which Riemann-Liouville definition form is the most widely used.

The fractional derivative Riemann-Liouville operator of the continuous function $f(t)$ is defined as

$$\left(D_{0^+}^{\gamma} f\right)(t) = \frac{1}{\Gamma(1-\gamma)} \frac{d}{dt} \int_0^t \frac{f(\tau)}{(t-\tau)^{\gamma}} d\tau \quad 0 \le \gamma \le 1 \tag{1}$$

If $s = t - \tau$, the integral transformation of equation (1) can be obtained

$$\left(D_{0^+}^{\gamma} f\right)(t) = I_{\gamma}(t) f(0) + \int_0^t I_{\gamma}(t-\tau) df(\tau) = \frac{1}{\Gamma(1+\gamma) t^{\gamma}} * df \tag{2}$$

Koeller (1984) proposed a constitutive equation for the Abel adhesive pot element with fractional derivative describing viscoelastic behaviour which can be written as

$$\sigma = D\eta^{\gamma}[\varepsilon(t)] = \eta I_{\gamma}(t) * d\varepsilon \tag{3}$$

Its creep compliance is

$$J(t) = \frac{1}{\eta} \phi(t) * \frac{t^{\gamma-1}}{\Gamma(\gamma)} = \frac{1}{\eta} \int_0^t \frac{t^r}{\Gamma(\gamma)} dt = \frac{1}{\eta \Gamma(\gamma+1)} t^r \tag{4}$$

where $\phi(t)$ is unit strain and $\phi(t) = \begin{cases} 1 \ , & t \ge 0 \\ 0 \ , & t < 0 \end{cases}$

Therefore, it can be seen from equation (2) that the fractional derivative is the convolution of the $I_r(t)$ of Abel with the Stielttjes of $f(t)$.

Creep function of viscoelastic polymer materials obeys Nutting formula (He, 2008) as follows

$$\varepsilon(t) = At^{\gamma}\sigma, 0 < \gamma < 1 \tag{5}$$

where A and γ are material parameters reflecting the material performance, which can be determined by test.

Its creep compliance is

$$J(t) = At^{\gamma}, 0 < \gamma < 1, \tag{6}$$

It can be seen from equation(4) and (6) that the creep flexibility of Abel clay pot is a power function of time as follows

$$A = \frac{1}{\eta\Gamma(\gamma+1)} \tag{7}$$

Equation (7) indicates A has certain physical significance, which is comprehensively affected by the viscosity coefficient of materials and the order of fractional derivatives.

2.2 Dynamic mechanical properties of fractional derivative viscoelastic constitutive model

For the fractional model the stress is proportional to the fractional derivative of the strain and the constitutive law is given by equation (8) and (9):

$$\tau(t) = \frac{1}{c_{\gamma}}\left(D_{0^+}^{\gamma} g\right)(t) \tag{8}$$

$$\varepsilon(t) = c_{\gamma}\left(I_{0^+}^{\gamma} \tau\right)(t) \tag{9}$$

where $\tau(t)$ is the shear stress, $g(t)$ is the shear strain and c_{γ} is constant value $c_{\gamma} = A_{\gamma}\Gamma(\gamma)$ (Fecarotti et al., 2012; Celauro et al., 2009), Γ is the Euler Gamma function and γ is the order of the fractional operator, $(I_{0+}^{\gamma}\sigma)(t) = \frac{1}{\Gamma(\gamma)}\int_0^t \frac{f(t)}{(t-\tau)^{1-\gamma}}\mathrm{d}\tau, f(t)$ is the continuous function of t.

The Fourier transform of equation (8) is obtained as follows:

$$\tilde{\tau}(\omega) = \frac{1}{c_{\gamma}}(i\omega)^{\alpha}\tilde{g}(\omega) \tag{10}$$

Further, the following equation can be obtained as follows:

$$G^*(\omega) = \frac{\tilde{\tau}(\omega)}{\tilde{g}(\omega)} = \frac{(i\omega)^{\gamma}}{c_{\gamma}} = c_{\gamma}^{-1}(i\omega)^{\gamma} \tag{11}$$

where $G^*(\omega) = G_1(\omega) + iG_2(\omega)$, in which, $G^*(i\omega)$ is complex shear modulus, $G_1(\omega)$ is storage modulus, and $G_2(\omega)$ is loss modulus.

The modulus of equation (11) can be obtained as follows:

$$G^* = \frac{\omega^{\gamma}}{A\gamma\Gamma(\gamma)} \tag{12}$$

3 Dynamic shear rheological tests

3.1 Experimental materials and preparation of modified asphalt

3.1.1 Asphalt

The asphalt used in this paper was produced in Jiangyin of China. The basic properties of asphalt are shown in Table 1.

Table 1 Physical properties of neat asphalt

Property	*Unit*	*Test result*	*Engineering requirements*	*Test method*
Penetration (100g, 5s, 25ºC)	0.1mm	74.80	60~80	T0604-2011
Ductility (5cm/min, 15ºC)	cm	≥100	≥100	T0605-2011
Softening point	ºC	47.5	≥43	T0606-2011

3.1.2 EVA particles

EVA is a ethylene-vinyl acetate copolymer. Vinyl acetate (VA) is generally between 5% and 40%. EVA and asphalt are naturally similar in solubility, so the compatibility between them is relatively good. EVA, as a modifier, has a good effect in improving the high-temperature stability and anti-deformation ability of asphalt.

Figure 1 Eva particles

In this paper, EVA particles were selected as asphalt modifier, which contained 15% VA content, with a diameter of about 0.5 cm. EVA particles were produced by YUCHENLONG engineering plastics company (see Figure 1). The physical properties of EVA plastic particles were shown in Table 2.

Table 2 Physical properties of EVA plastic particles(provided by manufacturer)

Property	*Unit*	*Test result*
VA mass fraction	%	15
Melt index	g/10min	8
Density(25°C)	g/cm^3	0.941
Fracture strength	kg/m^2	160
Ultimate elongation	%	700
Melting point	°C	110
Embrittlement temperature	°C	-70

3.1.3 Preparation of modified asphalt

The neat asphalt heated and melted in the oven and kept within 170 ± 5ºC for using. Then EVA modifier was kept as 0%, 3%, 6% and 9% by weight of the asphalt. And the speed of high-speed shear emulsifier (Shanghai FLUKO FA60) maintains at 4,000 rpm mixing and the modified asphalt was stirred for 1h at 170ºC to produce EVA modified asphalt after EVA was added to the asphalt.

3.2 Experimental program

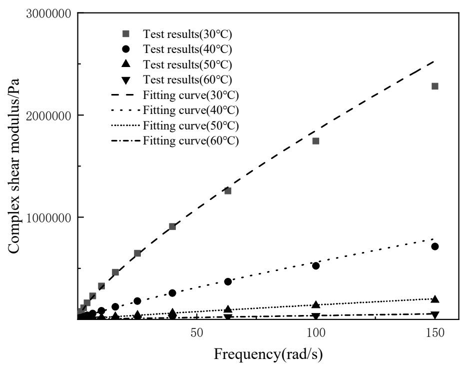
The DSR is primarily used to evaluate the rheological properties of polymer materials and is the standard instrument for evaluating the high temperature stability and fatigue durability of asphalt cementing materials in the US highway strategic research program (SHRP). The DSR testing is a advanced method to characterise the viscoelastic behaviour of asphalt mastic. The complex shear modulus (G^*) and the phase angle (δ) were calculated through its own software. The $G^*/\sin\delta$ was defined as the rutting factor and the high value of $G^*/\sin\delta$ meant the strong deformation resistance ability.

The frequency sweep tests in this paper were conducted on the specimens of 2,000 μm thick (at 30ºC) using 8mm diameter metal parallel plates and 1,000 μm thick (at 40ºC, 50ºC and 60ºC) using 25mm diameter metal parallel plates under controlled the frequency range from 0.1rad/s to 150rad/s. The tests were completed at the road material laboratory of Henan University of Urban Construction in April 2019. In order to ensure that all samples to be tested were within the range of small line viscoelasticity, the strain control index of the test was 2% because the dynamic shear rheological test was based on the assumption of line viscoelasticity.

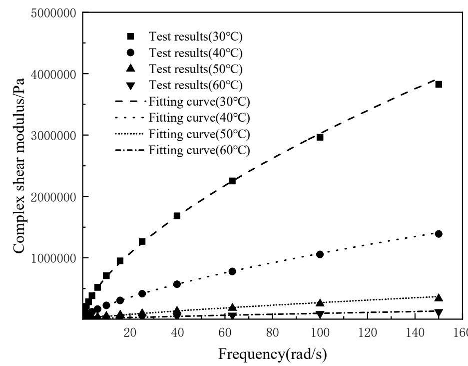
3.3 Results and discussion

The complex shear modulus tests results of dynamic frequency sweep tests of EVA modified asphalt were shown in Figure 2. It can be seen that the complex shear modulus gradually reduces with the increase of test temperature under the same EVA content. And the complex shear modulus increases with the increase of the test frequency at the same test temperature. In addition, the complex shear modulus of EVA modified asphalt gradually increases with the increase of EVA content. For example, under the test frequency of 150 rad/s, the complex shear modulus of modified asphalt with EVA content of 9% at 60ºC is 10.65 times that of asphalt with EVA content of 0. Therefore, the addition of EVA significantly improves the high-temperature deformation resistance of asphalt.

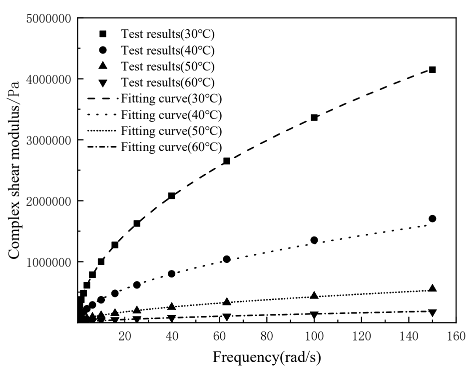
Figure 2 Complex shear modulus and fitting curves of eva modified asphalt, (a) 0% (b) 3% (c) 6% (d) 9%



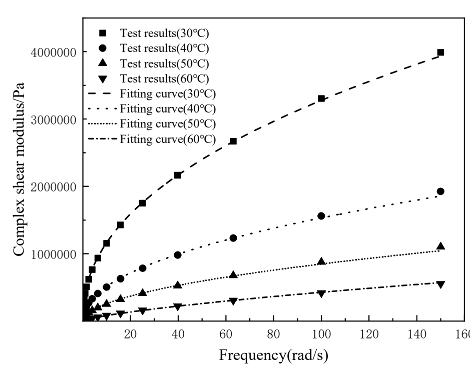
(a)



(b)



(c)

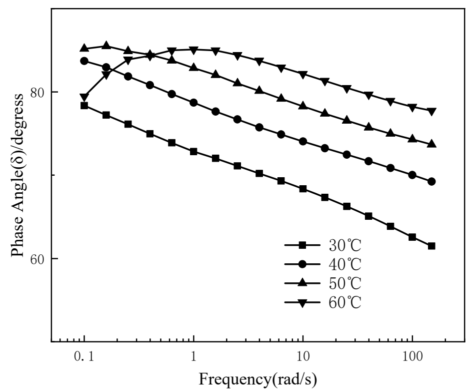


(d)

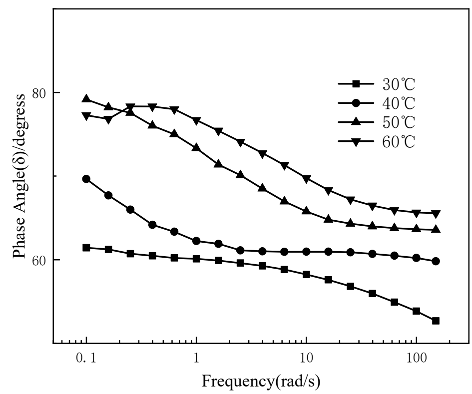
Figure 3 explains the effect of EVA on phase angle at different frequencies comparing with that of a neat asphalt binder. On the whole, with the increase of EVA content in modified asphalt, the phase angle of modified asphalt gradually decreases, which indicates that the addition of EVA increases the elasticity performance of modified asphalt and improves the high-temperature stability of modified asphalt. In addition, when the EVA content is 0% and 3%, the phase angle of modified asphalt gradually decreases with the increase of frequency. However, when the EVA content is 6% and 9%, the phase angle of modified asphalt is less affected by the change of frequency.

The phase angle of dynamic frequency scanning test of EVA modified asphalt is shown in Figure 3. It can be seen that the phase angle of asphalt mortar gradually decreases with the increase of EVA content in modified asphalt, which indicates that EVA addition increases the elasticity performance of modified asphalt and improves the stability of modified asphalt at high temperature. Moreover, when the EVA content is 0% and 3%, the phase angle of modified asphalt gradually decreases with the increase of frequency. However, when the EVA content is 6% and 9%, the phase angle of modified asphalt is less affected by the change of frequency.

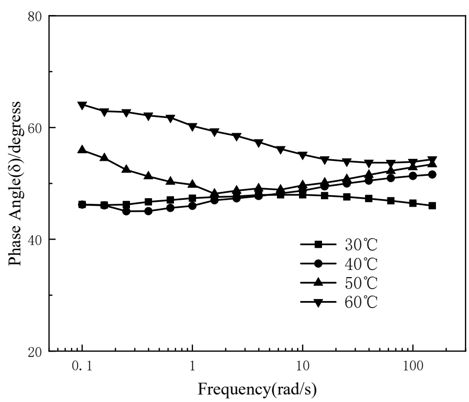
Figure 3 Phase angle under different loading frequencies, (a) 0% (b) 3% (c) 6% (d) 9%



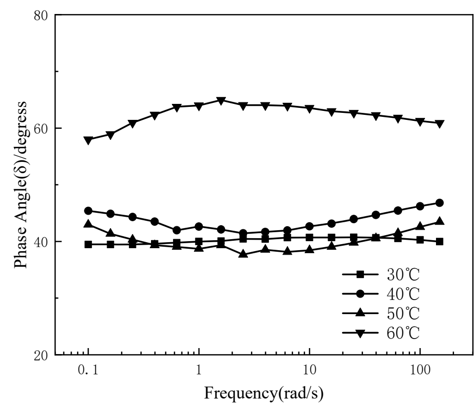
(a)



(b)

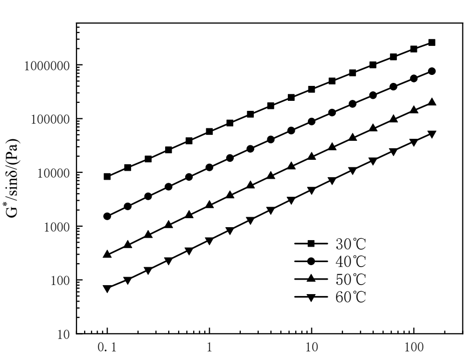


(c)

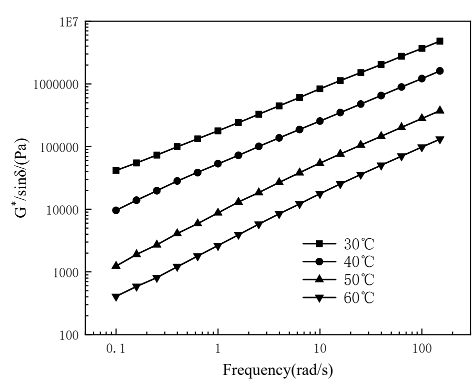


(d)

Figure 4 Rutting factor under different loading frequencies, (a) 0% (b) 3% (c) 6% (d) 9%

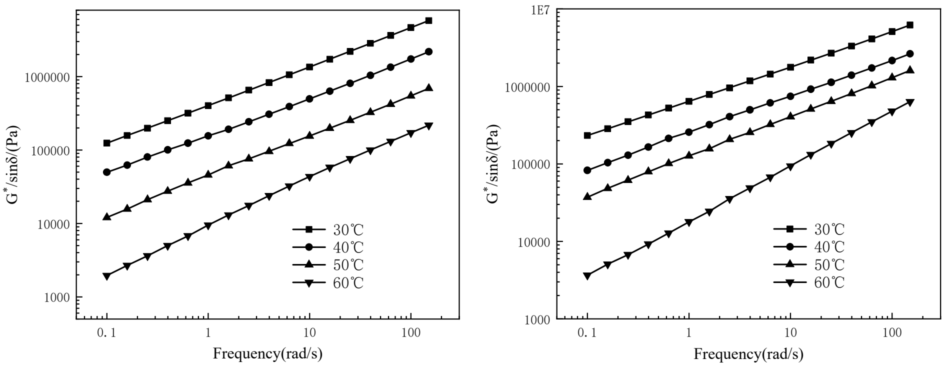


(a)



(b)

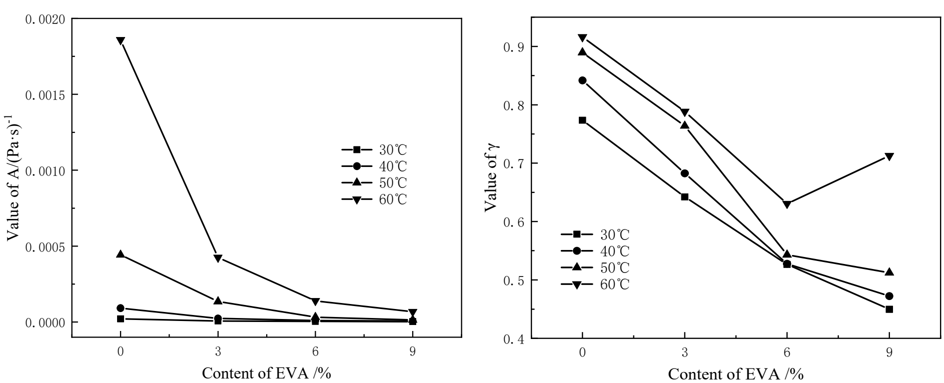
Figure 4 Rutting factor under different loading frequencies, (a) 0% (b) 3% (c) 6% (d) 9% (continued)



(c) (d)

According to Figure 4, the rutting factor goes up gradually with the increase of EVA content and frequency. Under the test frequency of 150 rad/s, the rut factor of modified asphalt with EVA content of 9% at 60ºC is 11.91 times that of asphalt with EVA content of 0.It indicates that the addition of EVA significantly improves the high temperature stability of modified asphalt.

Figure 5 The value of parameter A and γunder different EVA content



According to equation (12), there is

$$\lg\left|G^*(\omega)\right| = -\lg A\gamma\Gamma(\gamma) + \gamma\lg\gamma \tag{13}$$

Origin's linear fitting tool was used to perform power function fitting analysis on the relationship between complex shear modulus and frequency in dynamic frequency sweep tests of EVA modified asphalt based on equation (13). The fitting results were shown in the fitting curves in Figure 2 (the minimum adjusted complex decision coefficient was 0.9991).The parameters of the material can be calculated and shown in Figure 5. It can be seen that the value of A gradually decreases with the increase of EVA content, which is consistent with the material performance reflected in the equation (7). Moreover, the variation between the values of A at different temperatures becomes smaller and smaller with the increase of EVA content. The γ value decreases gradually with the increase of EVA content (except γ value at 60ºC and 9% EVA content).This indicates that the stronger the elasticity of the material, the smaller the γ value. Under the condition of the same EVA content, the γ value gradually increases with the increase of temperature, reflecting that the stronger the viscosity of the material is, the larger the γ value is. Therefore, we can call γ as viscoelastic factor to evaluate the viscoelastic behaviour of the material.

4 Summary and conclusions

a The integral transformation of fractional derivative Riemann-Liouville fractional derivative differential operator was carried out, and the relation between the fractional-order derivative Riemann-Liouville fractional derivative differential operator and the power function empirical creep equation was established. The physical meaning of power function parameters were clarified.

b The dynamic shear rheological tests show that the complex shear modulus and the rutting factor of the asphalt modified by EVA increases with the increase of EVA content and decreases with the increase of temperature. The results show that the high temperature deformation resistance of modified asphalt is improved by adding EVA.

c Based on the fractional derivative theory, the integral transformation of fractional-derivative Riemann-Liouville fractional derivative operator was carried out, and the relation between fractional-derivative operator and power function empirical creep equation was established. Origin software was used to perform power function fitting analysis on the relationship between complex shear modulus and frequency based on the dynamic shear rheological test results of EVA modified asphalt. The parameters A and γ which characterise the viscoelastic energy of the material were determined. The results show that the complex shear modulus of EVA modified asphalt has a good power function relationship with the test frequency.

References

Airey, G.D. (2002) ‘Rheological evaluation of ethylene vinyl acetate polymer modified bitumens’, *Construction Buildig. Materials*, Vol. 16, No. 8, pp.473–487.

Airey, G.D. (2003) ‘Rheological properties of styrene butadiene styrene polymer modified road bitumens’, *Fuel*, Vol. 82, No. 14, pp.1709–1719.

Alatas, T., Yilmaz, M., Kuloglu, N., et al. (2014) ‘Investigation of shear complex modulus and phase angle values of short- and long-term aged polymer modified bitumens at different frequencies’, *KSCE Journal of Civil Engineering*, Vol. 18, No. 7, pp.2093–2099.

Bagley, R.L. and Torvik, P.J. (1983) ‘A theoretical basis for the application of fractional calculus toviscoelasticity’, *J. Rheol*, Vol. 27, No. 3, pp.201–210.

Bagley, R.L. and Torvik, P.J. (1986) ‘On the fractional calculus model of visco elasticity behavior’, *J. Rheol*, Vol. 30, No. 19, pp.133–155.

Celauro, C., Di Paolam, M., Lo Presti, D., et al. (2009) ‘Modeling of the viscoelastic behavior of paving bitumen fractional derivatives’, *Meccanica dei Materiali e delle Strutture*, Vol. 1, No. 2, pp.38–51.

Fan, W., Ren, S., Liang, M., et al. (2017) 'Effect of EVA molecular structure on properties of EVA modified asphalt', *Journal of China University of Petroleum*, Vol. 41, No. 5, pp.159–168 (in Chinese).

Fecarotti, C., Celauro, C. and Pirrotta, A. (2012) 'Linear viscoelastic behaviour of bitumen via fractional models', *Social and Behavioural Sciences*, Vol. 53, No. 16, pp.450–461.

Gaputo, M. (1966) 'Linear models of dissipation whose Q is almost frequency indepedent', *Annalidi Geofisica*, Vol. 19, No. 19, pp.383–393.

García-Morales, M., Partal, P., Navarro, F.J. et al. (2006) 'Process rheokinetics and microstructure of recycled EVA/LDPE-modified bitumen', *Rheologica Acta*, Vol. 45, No. 4, pp.513–524.

Gemant, A. (1938) 'On fractional differences', *Philosophical Magazine*, Vol. 25, No. 9, pp.92–96.

He, P. (2008) *Mechanical Properties of High Polymer*, Version 2, Press of University of Science and Technology of China, Hefei, pp.104–105 (in Chinese).

Hu, J., Fang, C. and Zhou, S. (2015) 'Rheological properties of packaging-waste-polyethylene-modified asphalt', *Journal of Vinyl & Additive Technology*, Vol. 21, No. 3, pp.215–219.

Koeller, R.C. (1984) 'Applications of fractional calculus to the theory of viscoelasticity', *Journal of Applied Mechanics*, Vol. 51, No. 11, pp.229–307.

Kumar, P.R. and Anand, S. (2018) 'Enhancement of storage stability and rheological properties of polyethylene (PE) modified asphalt using cross linking and reactive polymer based additives', *Construction and Building Materials*, Vol. 188, No. 33, pp.772–780.

Qiu Sheng, H.U. and Gang, Z. (2015) 'The research on improvement of low temperature stability for diatomite modified asphalt mixture with PE particles', *International Journal of Engineering Research and Applications*, Vol. 5, No. 1, pp.01–05.

Shimizu, N. and Zhang, W. (1999) 'Fractional calculus approach to dynamic problem of viscoelastic materials', *JSME International Journal*, Vol. 42, No. 1, pp.827–830.

Yu, R., Fang, C., Liu, P., et al. (2015) 'Storage stability and rheological properties of asphalt modified with waste packaging polyethylene and organic montmorillonite', *Applied Clay Science*, Vol. 104, No. 21, pp.1–7.

Zhang, W., Shimizu, N. and Xu, H. (2002) 'Thermal effects of the viscoelastic materia1s described by fractional calculus constructive law', *The FIRST Asian Conference on Multibody Dynamics*, in Iwaki, Japan, pp.02–08.