

Viscoelastic Behavior and Constitutive Relation of Heavy Crude Oils

Hai-fei Liu, Yuan-yuan Xu, Hu Chen, Jian Zhang,* and Jing-yu Xu

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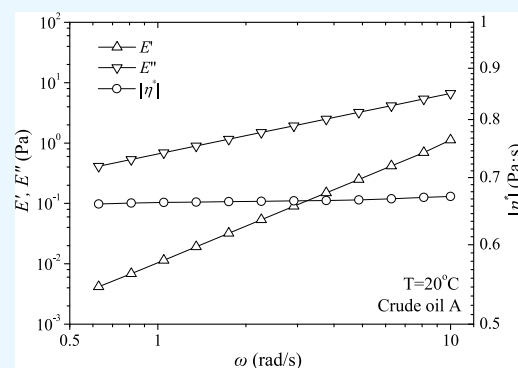
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ABSTRACT: Heavy crude oil exhibits very complex viscoelastic behaviors due to its complex composition of resins, asphaltenes, saturates, and aromatics. It has a great influence on oil production and transportation. In this work, the viscoelastic behaviors of three different heavy crude oils were measured using a rotational rheometer. In conclusion, all of these heavy crude oils display linear viscoelastic behaviors in the experimental range. The loss modulus (E'') of the three crude oils decreased as the experimental temperature increased, and the variation trends of the three crude oils were basically the same. However, the experimental temperature has almost no effect on the storage modulus (E'), which always retained a constant value of 0.4 Pa. Furthermore, the storage modulus (E') and loss modulus (E'') increase as the angular frequency increases. To describe the physical deformation characteristics of viscoelastic materials, the generalized Maxwell model and the fractional derivative Maxwell model are used to establish the constitutive relation of heavy crude oil. In conclusion, the generalized Maxwell model and the fractional derivative Maxwell model can predict the experimental results very well. All of the square of the correlation coefficient (R^2) values are greater than 0.95. However, the number of fitting parameters for the fractional derivative Maxwell model is less than that for the fourth-order generalized Maxwell model which can save the calculating time. Therefore, the fractional derivative Maxwell model is suggested to describe the viscoelastic behavior of heavy crude oil in industrial applications.



1. INTRODUCTION

Crude oil plays an important role in the world's energy structure. Heavy crude oil is one type of crude oil with a high density and viscosity.¹ The API value of heavy crude oil is always less than 20. Throughout the world, the reserves of heavy crude oil are more than twice those of conventional light and medium crude oil. The mining of heavy crude oil is becoming more important to supply fuel for economic development. However, due to the different components of saturates, aromatics, resins, and asphaltenes, most heavy crude oils demonstrate some complex rheological behaviors, such as shear thinning, thixotropy, and viscoelasticity.^{2,3} The rheological behaviors of heavy crude oil are very complex and are different from those of light crude oil. Therefore, they have a great influence on the production of heavy crude oil, especially on the processes of mining and transportation.⁴

The viscoelastic behavior of heavy crude oil is one of the most important rheological characteristics that can affect the shutdown and restart of sections of pipeline transportation.^{5,6} According to the definition of yield stress, the fluid will return to its original state due to the elastic characteristic if the stress is removed and less than the yield stress.⁷ The viscoelastic behavior of heavy crude oil is very complex because of the different components, and the environment temperature is changing, which can greatly affect the viscoelastic behavior.⁸

To study the viscoelasticity of heavy crude oil, experimental tests and theoretical analyses are always used in the literature.⁹ Creep-recovery tests and dynamic oscillatory shear tests are two classical experimental methods in the development of rheological test technology. The creep-recovery behavior of heavy crude oil is often described by mechanical analogy models, where the fitted parameters can be used to analyze the structural evolution under a constant stress.¹⁰ In this process, the yield stress is an important parameter that can obviously affect the creep behavior. It can be divided into three stages, namely, instant elastic deformation, decelerated creep, and constant rate creep, when the loading stress is less than the yield stress. The accelerated creep process occurs after these three stages when the loading stress is larger than the yield stress. The dynamic oscillatory shear test method can be used to study the viscoelastic characteristics over a wide range of shear frequencies and amplitudes.¹¹ The test consequence of the storage modulus and loss modulus can be used to directly describe the elastic and viscous behaviors of heavy crude oil.¹²

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The viscoelastic characteristics of crude oil have been researched for a long time, and most of them have focused on waxy crude oils.¹³ Guo et al.¹⁴ gave one viscoelastic-thixotropic model with nine parameters based on the principle of mechanical comparison, which can be used to describe the viscoelastic behaviors of wax crude oils. Strelets et al.¹⁵ systematically researched the effect of enhanced oil recovery on the rheological properties. This shows that heavy crude oil is a viscoelastic glass-forming liquid that can exhibit non-Newtonian behavior, and the temperature has a great influence on the rheological behaviors. Moreover, the effects of shear stress and oscillation on the structural characteristics of gelled crude oil have been studied through rheological experiments. In the results, the linear viscoelastic region and yielding can be determined by the critical linear shear stress or strain and yield stress or strain, respectively. To improve pipeline transportation, some startup flow experiments have been designed to research the yield stress or viscoelastic behaviors. Additionally, the effect of emulsifying water on the viscoelastic characteristics of crude oil has been studied.¹⁶ The droplet size and distribution have a great influence on the viscoelastic parameters, including the storage modulus and loss modulus.¹⁷

To understand the viscoelastic behavior of heavy crude oil more clearly, three different heavy crude oils from different oilfields or wells are chosen to study their viscoelastic behaviors. The components of these oil samples are very different. A series of rheological experiments including the oscillatory shear flow are designed to measure the relevant rheological behaviors. Furthermore, the analogy method is used to establish the viscoelastic constitutive relation of heavy crude oil. Two chosen constitutive relations, namely, the generalized Maxwell model and fractional derivative Maxwell model, are used to analyze the experimental results of viscoelastic characteristics. It is useful to accurately analyze the viscoelastic behavior of heavy crude oil.

2. VISCOELASTICITY CONSTITUTIVE RELATIONS

To describe the physical deformation characteristics of viscoelastic materials, the analogy method is always used to establish the constitutive relation.¹⁸ The Hooke spring and Newtonian dashpot are two basic units used to describe these behaviors of elasticity and viscosity, respectively.¹⁹ Moreover, the Maxwell model and Kelvin model are introduced to describe the viscoelastic characteristics.²⁰ The Maxwell model consists of a spring and a dashpot in series, and the Kelvin model consists of a spring and a dashpot in parallel. Physically, the Maxwell model is always used to describe the stress relaxation phenomenon of a viscoelastic material under static conditions, the Kelvin model is always used to describe the creep phenomenon, and its storage modulus is independent of the stress frequency. Therefore, the Maxwell model and Kelvin model cannot be used to accurately describe the viscoelasticity characteristics of crude oil. Two classic models, the generalized Maxwell model and fractional derivative model, have been developed to solve this question.^{21,22}

2.1. Generalized Maxwell Model. The generalized Maxwell model consists of some Maxwell models in parallel, which is shown in Figure 1. The elastic modulus E and viscous modulus η of every Maxwell model are different.

The relaxation modulus of the generalized Maxwell model can be calculated as follows.²³

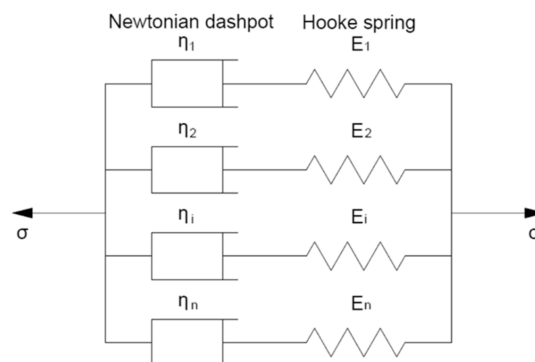


Figure 1. Generalized Maxwell model.

$$E(t) = \sum_{i=1}^n E_i \exp\left(-\frac{E_i t}{\eta_i}\right) \quad (1)$$

Here, n is the model order of the generalized Maxwell model. The storage modulus and loss modulus of the generalized Maxwell model can be obtained by the Fourier transformation.

$$E' = \sum_{i=1}^n E_i \frac{\omega^2 \tau_i^2}{1 + \omega^2 \tau_i^2} \quad (2)$$

$$E'' = \sum_{i=1}^n E_i \frac{\omega \tau_i}{1 + \omega^2 \tau_i^2} \quad (3)$$

Here, τ_i is the relaxation time, $\tau_i = \eta_i/E_i$.

2.2. Fractional Derivative Maxwell Model. Compared to the traditional viscoelastic model, the Abel dashpot is applied to replace the Newtonian dashpot in the fractional derivative model. The constitutive relation of the Abel dashpot is displayed below.

$$\sigma(t) = E \tau^\alpha D^\alpha \varepsilon(t) = E^{1-\alpha} \eta^\alpha D^\alpha \varepsilon(t) \quad (4)$$

Here, E is the elastic modulus; η is the viscosity coefficient; $\tau = \eta/E$ is the relaxation time; and D^α is the fractional derivative operator.

The fractional derivative Maxwell model consists of an Abel dashpot and a Hooke spring, which is shown in Figure 2. Therefore, the constitutive relation of the fraction Maxwell model can be shown in eq 2.

$$\sigma(t) = \frac{E_1 E \tau^\alpha D^\alpha}{E_1 + E \tau^\alpha D^\alpha} \varepsilon(t) \quad (5)$$

Here, E_1 is the elastic modulus of the Hooke spring.

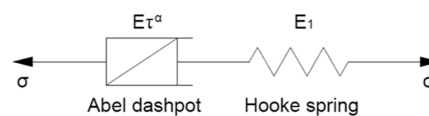


Figure 2. Fractional derivative Maxwell model.

When applying the sinusoidal alternating stress in this model, the corresponding strain can be calculated as shown in eq 3. Therefore, the relation between stress and strain can be obtained.

$$\varepsilon(t) = \varepsilon_0 \exp(i\omega t) \quad (6)$$

$$\sigma(\omega) = \frac{E_1 E \tau^\alpha (i\omega)^\alpha}{E_1 + E \tau^\alpha (i\omega)^\alpha} \varepsilon(\omega) \quad (7)$$

$$\sigma(\omega) = \frac{E_1 E \tau^\alpha \omega^\alpha \left(\cos \frac{\alpha\pi}{2} + i \sin \frac{\alpha\pi}{2} \right)}{E_1 + E \tau^\alpha \omega^\alpha \left(\cos \frac{\alpha\pi}{2} + i \sin \frac{\alpha\pi}{2} \right)} \varepsilon(\omega) \quad (8)$$

According to the definition of the complex modulus E^* , the following formulas to calculate the storage modulus E' and loss modulus E'' are obtained.

$$E^* = E' + iE'' = \frac{\sigma(\omega)}{\varepsilon(\omega)} \quad (9)$$

$$E' = \frac{E_1 E \tau^\alpha \omega^\alpha \left(E_1 \cos \frac{\alpha\pi}{2} + E \tau^\alpha \omega^\alpha \right)}{E_1^2 + 2E_1 E \tau^\alpha \omega^\alpha \cos \frac{\alpha\pi}{2} + E^2 \tau^{2\alpha} \omega^{2\alpha}} \quad (10)$$

$$E'' = \frac{E_1^2 E \tau^\alpha \omega^\alpha \sin \frac{\alpha\pi}{2}}{E_1^2 + 2E_1 E \tau^\alpha \omega^\alpha \cos \frac{\alpha\pi}{2} + E^2 \tau^{2\alpha} \omega^{2\alpha}} \quad (11)$$

3. EXPERIMENTAL SECTION

3.1. Experimental Methods. The viscoelastic characteristics and other rheological behaviors of heavy crude oil were measured using a HAAKE RS6000 rotational rheometer. A coaxial cylinder sensor system Z38 with a cup Z43 was chosen in this work. The diameter of rotor Z38 is 38 mm, and the inner diameter of cup Z43 is 43 mm. Therefore, the gap between the rotor and cup is 2.5 mm. The shear rate range of this rheometer is 0.001–1500 s^{-1} , and the viscosity range is 0.5– 10^6 mPas. A variety of temperature control units are available to handle experimental temperatures from 0 to 100 °C with an accuracy of 0.1 °C. In this work, the experimental temperature was controlled in the range from 20 to 70 °C.

The viscoelastic behaviors of heavy crude oil were measured using the small amplitude oscillatory shear measurement method. The shear stress control test (CS) model was used in this study. In the CS model, the shear stress is assigned an amplitude and an angular frequency periodicity, and the corresponding strain is measured. A shear stress sweep with a constant frequency was first performed to obtain the linear viscoelastic region of crude oil. Then, a frequency sweep was performed to systematically measure the viscoelastic characteristics of a series of heavy crude oils.

3.2. Materials. Three different types of heavy crude oils were chosen as the research samples to improve the range of application of this work. Crude oils A and B were produced from two different blocks in the Bo-hai offshore oilfield, and crude oil C was produced in the Sui-zhong onshore oilfield in China. All of these crude oils display non-Newtonian fluid characteristics, such as shear thinning and thixotropy. The physical properties, including density, viscosity, and saturate, aromatic, resin, and asphaltene (SARA) analysis, of these experimental crude oils are given in Table 1. This shows that the physical properties of these crude oils are greatly different. The density and viscosity of crude oil C are much higher than those of the other two crude oils. Some rheological characteristics of crude oils B and C can be found in the study by Zhang et al. (2017).³ Also, the flow curve for crude oil A at different temperatures is shown in the Appendix.

Table 1. Physical Properties of the Experimental Oils ($T = 30$ °C, $P = 101.325$ kPa, $\dot{\gamma} = 10$ s^{-1})

oil types	density (kg/m ³)	viscosity (mPas)	SARA analysis (wt %)			
			saturates	aromatics	resins	asphaltenes
crude oil A	933	336	58.6	20.4	19.2	1.8
crude oil B	920	242	52.7	27.3	18.4	1.6
crude oil C	955	5439	33.2	26.2	37.2	3.4

4. RESULTS AND DISCUSSION

4.1. Viscoelastic Behaviors of Heavy Crude Oils. Heavy crude oil exhibits very complex viscoelastic behaviors due to the complex composition of resins, asphaltenes, saturates, and aromatics. The different contents of these components have a great influence on the rheological characteristics. In this work, the viscoelastic behaviors of three different heavy crude oils were measured systematically. First, the shear stress sweep measurement was performed to confirm the linear range of the viscoelasticity. As reported by Behzadfar and Hatzikiriakos,²⁴ preferably high frequencies should be chosen to ensure the linearity at lower frequencies as well. The maximum value of the sweep frequency was 10 rad/s in this work.

The shear stress sweep measurement results of crude oil A at different temperatures are shown in Figure 3. The angular

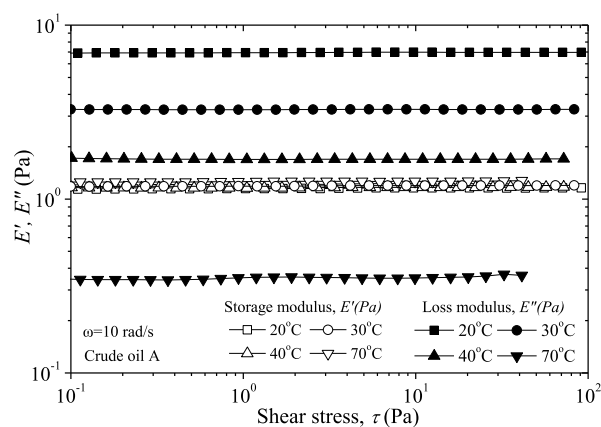


Figure 3. Shear stress sweep measurement results at different temperatures.

frequency was maintained at a constant value of 10 rad/s, which is the maximum value of the sweep frequency in this work. The experimental temperatures were 20, 30, 40, and 70 °C. It shows that the storage modulus E' and the loss modulus E'' are always kept at a constant value as the shear stress increases in the experimental range. In other words, the heavy crude oil sample displays linear viscoelastic behaviors in the experimental range. The effect of temperature on the storage modulus E' and loss modulus E'' of crude oils A, B, and C is shown in Figure 4. We can see that the loss modulus E'' of the three crude oils decreases as the experimental temperature increases. These variation trends of the three crude oils are basically the same. In contrast, the storage modulus E' is always kept at a constant value when the experimental temperature changes. Furthermore, the value of the storage modulus E' of these three heavy crude oils is almost the same.

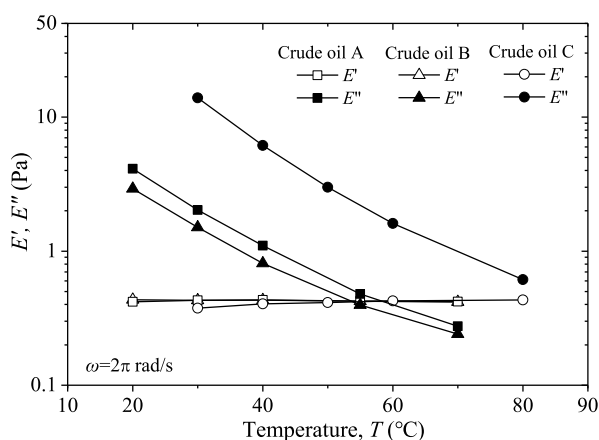


Figure 4. Effect of the temperature on the viscoelastic characteristics of crude oils.

Frequency sweep measurements were performed to study the linear viscoelastic characteristics of these heavy crude oils.^{25,26} One of the experimental results is shown in Figure 5.

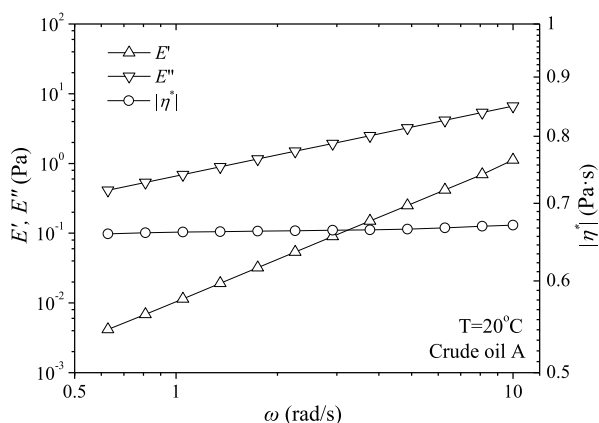


Figure 5. Frequency sweep measurement results in the linear viscoelastic region.

The shear stress was 1 Pa, which is maintained in the linear viscoelastic region. This shows that the storage modulus E' and loss modulus E'' continue to increase as the angular frequency increases. The storage modulus E' is less than the corresponding loss modulus E'' in the experimental range of the shear angular frequency from 0.6 to 10 rad/s. The relation between the storage modulus E' and angular frequency and the relation between the loss modulus E'' and angular frequency are the same for the index law. Additionally, the coefficient of complex viscosity was always kept at a constant value of 0.66 Pas. It demonstrates that the sample displays a linear viscoelastic behavior when the shear angular frequency is less than 10 rad/s. As a research consequence in the literature, the density can be used to evaluate the viscoelastic characteristics of crude oil. Therefore, the relation between the modulus and density in this work is shown in Figure 6. This shows that the storage modulus E' has little to do with the density. However, the relation between the loss modulus and density can be described by the index increase.

4.2. Analysis of Constitutive Relations. Following the experimental results, we can see that the viscoelastic behavior of heavy crude oil is very complex. The relation between the modulus and frequency cannot be described by a simple

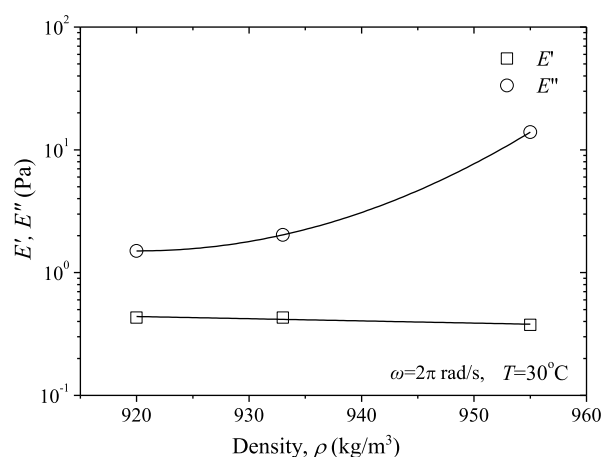


Figure 6. Effect of density on the viscoelastic characteristics of crude oils.

constitutive relation, such as the Maxwell model or Kelvin model. In this section, the feasibility and accuracy of the generalized Maxwell model and the fractional derivative Maxwell model used to analyze the viscoelastic behavior of heavy crude oil are studied. In this work, the fourth-order generalized Maxwell model was chosen to describe the viscoelastic characteristics of crude oil because it has a great advantage in terms of efficiency and accuracy. All the fitting parameters of the two models are shown in Tables 2 and 3,

Table 2. Parameters of the Fourth-Order Generalized Maxwell Model

	crude oil A	crude oil B	crude oil C
E_1	8.2913	4.2614	356.2415
E_2	2.5124	1.6822	272.1226
E_3	0.2465	1.5537	12.4178
E_4	0.0071	0.0213	0.0064
τ_1	0.0278	0.0401	0.0039
τ_2	0.0461	0.0503	0.0022
τ_3	0.0019	0.0072	0.0142
τ_4	7.5615	7.5269	6.5872

Table 3. Parameters of the Fractional Derivative Maxwell Model

	crude oil A	crude oil B	crude oil C
E_1	14.8757	9.9606	625.8378
E	0.7349	0.2875	0.1234
τ	0.4941	1.0887	0.8374
α	0.9501	0.9202	0.9986

respectively. The minimum and maximum values of the relaxation time (τ_i) and spring constants (E_i) in the generalized Maxwell model are limited following those in the literature studies.^{27,28} As a constraint in the optimum fitting calculation, the fractional order (α) in the fractional Maxwell model is between 0 and 1.^{29,30}

In the data analysis, the Levenberg–Marquardt (LM) method and the general global optimization method were used in the process of data fitting. The square of the correlation coefficient (R^2) was used to measure the fitting results of the generalized Maxwell model and fractional derivative Maxwell model. The value of R^2 is in the range of

0–1, and the fitting result is better when the value of R^2 is close to 1. Furthermore, all of the parameters in the generalized Maxwell model and fractional derivative Maxwell model were limited to a reasonable range based on the physical analysis.

$$R_{xy} = \frac{\sum_{i=1}^n (x_i - \bar{x})(y_i - \bar{y})}{\sqrt{\sum_{i=1}^n (x_i - \bar{x})^2} \sqrt{\sum_{i=1}^n (y_i - \bar{y})^2}} \quad (12)$$

The prediction consequences of the storage modulus and loss modulus of these three different heavy crude oils are shown in Figures 7 and 8, respectively. This shows that the generalized Maxwell model and the fractional derivative

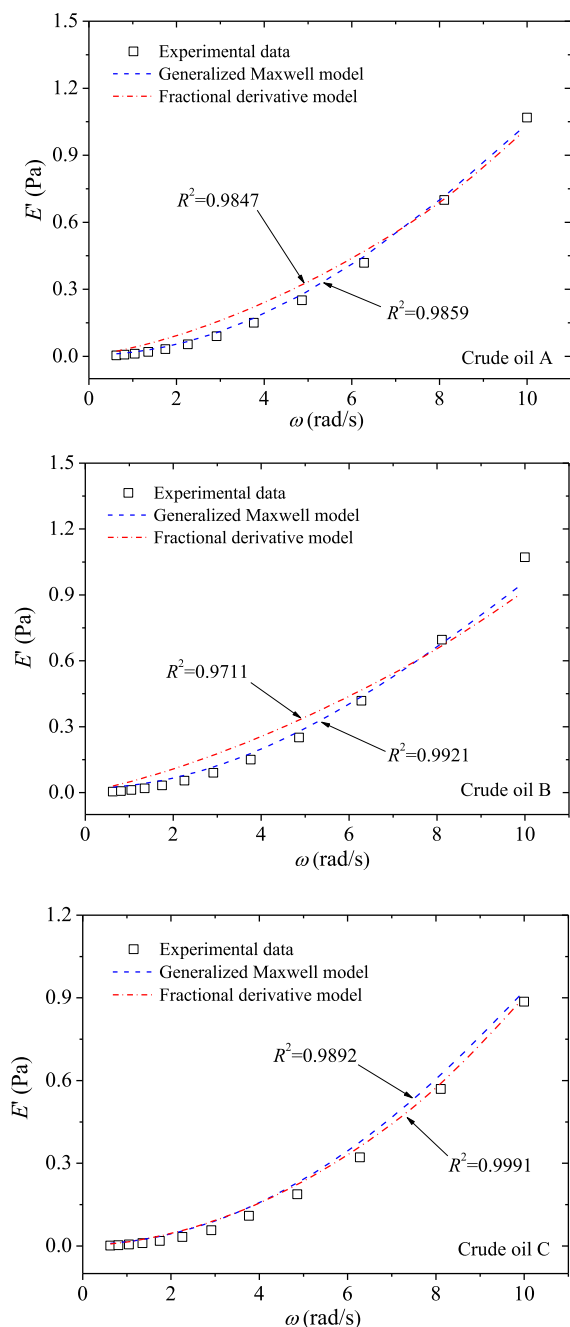


Figure 7. Prediction consequence of the storage modulus of different heavy crude oils.

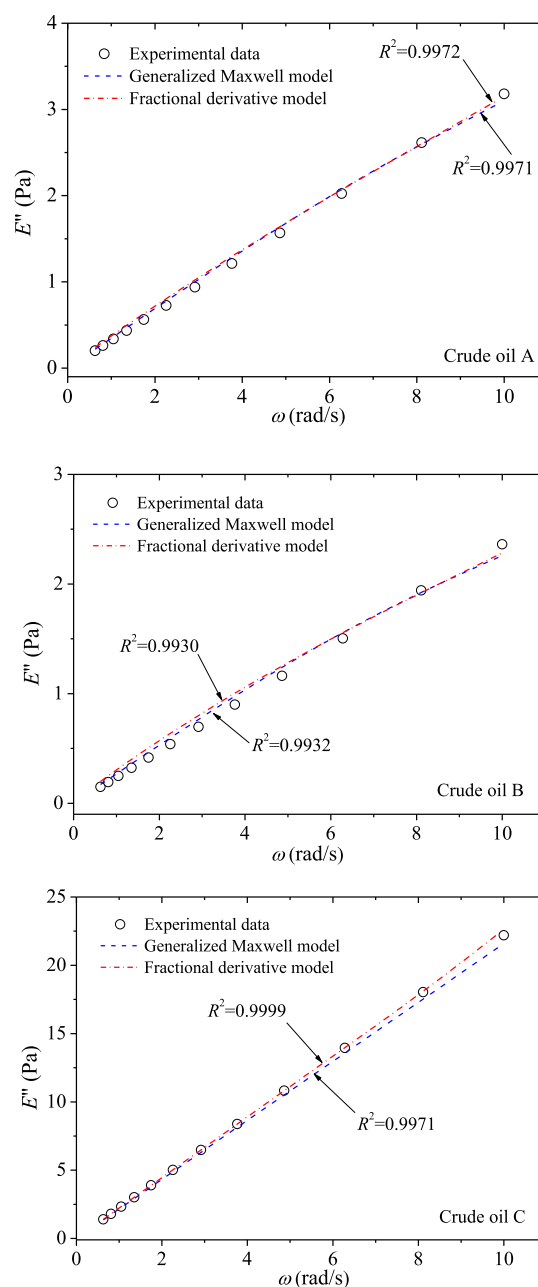


Figure 8. Prediction consequence of the loss modulus of different heavy crude oils.

Maxwell model can predict the experimental results very well. All of the R^2 values are greater than 0.95. For crude oil A, the R^2 values of the fractional derivative Maxwell model are 0.9847 and 0.9972 for the storage modulus and loss modulus, respectively, which are about the same as the fourth-order generalized Maxwell model. In other words, the prediction accuracy of the fractional derivative Maxwell model and the fourth-order generalized Maxwell model is basically the same. Furthermore, the same results can be obtained from crude oils B and C.

5. CONCLUSIONS

Heavy crude oil exhibits very complex viscoelastic behaviors due to the complex composition of resins, asphaltenes, saturates, and aromatics. The different contents of these

compositions have a great influence on the rheological characteristics. In this work, the viscoelastic behaviors of three different heavy crude oils were measured using a rotational rheometer with two methods, shear stress sweep and frequency sweep. In conclusion, all of these heavy crude oils display linear viscoelastic behaviors in the experimental range. The loss modulus (E'') of the three crude oils decreased as the experimental temperature increased, and the variation trends of the three crude oils were basically the same. However, the experimental temperature has almost no effect on the storage modulus (E'), which was always kept at a constant value of 0.4 Pa. Furthermore, the storage modulus (E') and loss modulus (E'') increase as the angular frequency increases.

The experimental results show that the relation between the modulus and frequency cannot be described by a simple constitutive relation, such as the Maxwell model or Kelvin model. To describe the physical deformation characteristics of viscoelastic materials, the generalized Maxwell model and the fractional derivative Maxwell model are used to establish the constitutive relation of heavy crude oil. In conclusion, the generalized Maxwell model and the fractional derivative Maxwell model can predict the experimental results very well. All of the R^2 values are greater than 0.95. For crude oil C, the prediction accuracy of the fractional derivative Maxwell model is slightly better than that of the generalized Maxwell model which can be found in some literature studies.^{29,31} Then, the number of fitting parameters for the fractional derivative Maxwell model is four which is less than that for the fourth-order generalized Maxwell model which can save the calculating time. Therefore, the fractional derivative Maxwell model is suggested to describe the viscoelastic behavior of heavy crude oil in industrial applications.

APPENDIX

Flow curves for crude oil A at different temperatures (Figure A1).

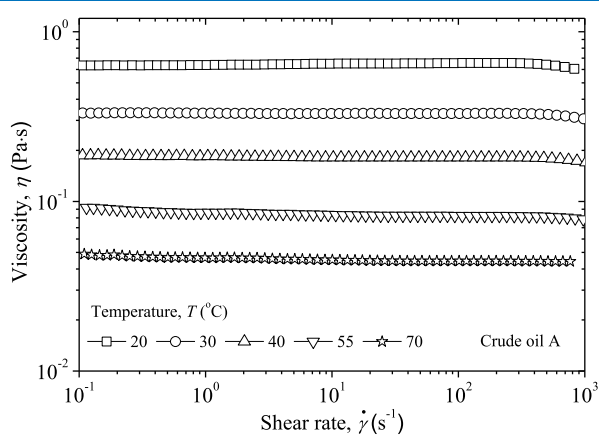


Figure A1. Flow curves for crude oil A at different temperatures (20, 30, 40, 55, and 70 °C).

AUTHOR INFORMATION

Corresponding Author

Jian Zhang – State Key Laboratory of Technologies in Space Cryogenic Propellants, Beijing 100028, China; Institute of Mechanics, Chinese Academy of Sciences, Beijing 100190,

China; orcid.org/0000-0002-0345-1443;

Email: zhangjian@imech.ac.cn

Authors

Hai-fei Liu – State Key Laboratory of Technologies in Space Cryogenic Propellants, Beijing 100028, China; Beijing Institute of Space Launch Technology, Beijing 100076, China

Yuan-yuan Xu – State Key Laboratory of Technologies in Space Cryogenic Propellants, Beijing 100028, China

Hu Chen – Institute of Mechanics, Chinese Academy of Sciences, Beijing 100190, China

Jing-yu Xu – Institute of Mechanics, Chinese Academy of Sciences, Beijing 100190, China

Complete contact information is available at:

<https://pubs.acs.org/10.1021/acsomega.2c02234>

Author Contributions

The manuscript was written through contributions of all authors. All authors have given approval to the final version of the manuscript.

Notes

The authors declare no competing financial interest.

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