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Research paper

The application of fractional derivatives through Riemannliouville approach to xanthan gum viscoelasticity

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Abstract

Fractional derivatives are derivative with non-integer order, one of which is used for mathematical modeling of viscoelasticity. In this research, the fractional derivative was used to obtain a mathematical model of viscoelasticity. The method used was a fractional derivative through the Riemann-Liouville approach. The mathematical model of viscoelasticity obtained was a complex modulus consisting of storage and loss modulus. This model was applied to xanthan gum concentrate solution 0.5%, 1.0%, 2.0%, 3.0%, and 4.0% with simplified model parameters. The results obtained that the storage and loss modulus increased with increasing concentration of the solution. In addition, the modulus storage was always greater than the modulus loss for all concentrations of the solution. This suggests that the elastic properties of the xanthan gum solution are more dominant than their viscosity properties for all concentrations. Therefore, the viscoelasticity model using Riemann-Liouville fractional derivatives has a good ability to investigate the viscoelasticity behavior of all xanthan gum concentrations.

Keywords: *Fractional Derivatives; Riemann-Liouville; Viscoelasticity; Xanthan Gum.*

1. Introduction

The derivation of a function is usually the order of positive integer numbers. Assume the function $y(x)$, then it can be determined the 1st, 2nd, 3rd, and so on of $y(x)$. Generalization of this concept is how to determine the derivatives to-α of a function where $\alpha \in \mathbf{R}$. When $D_x^n y(x)$ is a notation derived to-n of the function $y(x)$ with $n \in \mathbf{N}$, so the generalization of the form is notated by $D_x^\alpha y(x)$ as an instance to-α of the function $y(x)$ where $\alpha \in \mathbf{R}$. This concept was later known as the fractional derivative [12].

Many mathematicians such as Euler, Lacroix, Liouville, Riemann, Grünwald, Letnikov, Caputo, Weyl, Riesz, Laurent, Hadamard, Chen, and Marchaud contributed in developing the fractional derivative concept. Thus, the fractional derivative approaches are quite numerous and varied but still proved to be mutually equivalent. Today the application of fractional derivatives is used in various fields such as mathematics, physics, and biology, for example, the Schrödinger equation [11]; the diffusion equation [4], the fractional dynamic system has the control theory [6], electrochemistry and fluid flow, the fractional neuron model in biology, and viscoelasticity.

Viscoelasticity is a material property that exhibits viscous (e.g. dashpot) and elastic (e.g. spring) viscous characteristics when deformed [1]. When the stress (force) is removed in viscoelastic material, the material cannot directly return to its original state. One material that meets viscoelastic properties is xanthan gum. Xanthan gum is a high molecular weight polysaccharide produced by fermenting pure cultures from carbohydrates by Xanthomonas campestris bacteria, then purified with alcohol, dried and milled [2]. At low concentrations, the xanthan gum solution showed a high viscosity compared to other polysaccharide solutions. So this trait makes it a highly effective thickener and stabilizer. The xanthan gum solution is highly pseudoplastic, meaning that as the strain rate increases the viscosity decreases and vice versa. The xanthan gum solution is highly resistant to pH variation, which is stable in both acidic and alkaline conditions [14].

Based on research [15] we have investigated the dynamic viscoelastic properties of xanthan gum solution with different concentrations measured at various strains, and linear viscoelasticity with relatively small strain that measured by angular frequency. The tool used in the study was a rheometer controlled by a strain (Advanced Rheometric Expansion System (ARES), Rheometric Scientific, and Piscataway, NJ, USA). The xanthan gum samples used in the study were xanthan gum commercially available from Sigma-Aldrich Corporation (St. Louis, MO, USA), with concentrations of 1.0%, 2.0%, 3.0%, and 4.0%. The relationship of storage and loss modulus with the angular frequency in the linear viscoelastic xanthan gum solution can also be explained quantitatively by the power-law equation. As expected, the measured experimental data on the power-law equations adequately represent the linear viscoelastic of all solution concentrations. Then the least squares method was used to determine the linear fractional linear model of fractional viscolysis parameters from storage measurement and modulus loss. Further studies [5], still with the Riemann-Liouville fractional derivative model to investigate viscoelasticity in xanthan gum, with 0.5%, 1.0%, 2.0%, and 4.0%.

Based on the background of the problem, a study was conducted on fractional derivatives which were used to model the viscoelasticity and applies the model to xanthan gum solution with concentrations of 0.5%, 1.0%, 2.0%, 3.0%, and 4.0%, without interfering with other experimental result data.

2. Materials and methods

Fractional calculus is a term used to denote calculus by non-integer order, this concept appears alongside integer calculus independently initiated by Isaac Newton and Gottfried Leibniz [7], the calculus here refers to derivatives and integrals. Here is a fractional derivative through the Riemann-Liouville approach [8]:

$$({}_aD_x^\alpha f)(x)=\frac{1}{\Gamma(n-\alpha)}\frac{d^n}{dx^n}\int_a^x (x-y)^{n-\alpha-1} f(y)dy \tag{1}$$

Where $n-1\le\alpha<n$.
The following is an experimental data table with a simplified parameter model derived from the study in [15], [5], where G' is storage modulus and G" is loss modulus, and k1, k2, α_1, and α_2 are simplified parameters.

Table 1: Parameter Model of Fractional Derivatives Model

Concentration	G' and G"	k1	k2	α_1	α_2
0.5%	G'	17.6	-15.8	0.3444	0.3430
	G"	17.6	-15.8	0.1622	0.1760
1.0%	G'	37	190	0.1838	0.1842
	G"	-289	603	0.1313	0.1318
2.0%	G'	-347	655	0.2839	0.2304
	G"	-94	484	0.1380	0.1382
3.0%	G'	-530	1110	0.2413	0.1905
	G"	-95	814	0.1162	0.1169
4.0%	G'	-407	1249	0.2296	0.1646
	G"	-3992	5035	0.1123	0.1123

Before obtaining a mathematical model of viscoelasticity through Riemann-Liouville fractional derivatives, the following is a Fourier transformation for Riemann-Liouville fractional derivative with an infinite negative threshold and $n-1\le\alpha<n$ [9]:

$$F\left\{({}_{-\infty}D_x^\alpha f)(x)\right\}=F\left\{\frac{1}{\Gamma(n-\alpha)}\frac{d^n}{dx^n}\int_{-\infty}^x (x-y)^{n-\alpha-1} f(y)dy\right\} \tag{2}$$

For example $x-y=r$ then $y=x-r$ and $dy=-dr$, so for $y=-\infty$ then $r=\infty$ and $y=x$ and $r=0$, thus obtained:

$$\begin{aligned}F\left\{({}_{-\infty}D_x^\alpha f)(x)\right\}&=F\left\{\frac{1}{\Gamma(n-\alpha)}\frac{d^n}{dx^n}\int_\infty^0 r^{n-\alpha-1} f(x-r)(-dr)\right\}\\&=F\left\{\frac{1}{\Gamma(n-\alpha)}\frac{d^n}{dx^n}\int_0^\infty r^{n-\alpha-1} f(x-r)dr\right\}.\end{aligned}$$

Defined $H(r)$ which is a notation of Heaviside function that is 1 for $r>0$ and 0 for $r<0$ [13], then:

$$F\left\{({}_{-\infty}D_x^\alpha f)(x)\right\}=F\left\{\frac{1}{\Gamma(n-\alpha)}\frac{d^n}{dx^n}\int_{-\infty}^\infty H(r) r^{n-\alpha-1} f(x-r)dr\right\}.$$

Based on convolution in Fourier transform [3], hence:

$$\begin{aligned}F\left\{({}_{-\infty}D_x^\alpha f)(x)\right\}&=F\left\{\frac{d^n}{dx^n}\left[\frac{H(x)x^{n-\alpha-1}}{\Gamma(n-\alpha)}*f(x)\right]\right\}\\&=F\left\{\frac{d^n}{dx^n}\frac{H(x)x^{n-\alpha-1}}{\Gamma(n-\alpha)}*f(x)\right\}\\&=F\left\{\frac{d^n}{dx^n}\frac{H(x)x^{n-\alpha-1}}{\Gamma(n-\alpha)}\right\}F\{f(x)\}.\end{aligned}$$

The Fourier transform of the first term in the convolution above can be calculated using the definition and the result is [9]:

$$F\left\{\frac{H(x)x^{\gamma-1}}{\Gamma(\gamma)}\right\}=(i\omega)^\gamma,$$

For $\gamma>0$. So also obtained:

$$F\left\{({}_{-\infty}D_x^\alpha f)(x)\right\}=(i\omega)^\alpha \hat{f}(\omega). \tag{3}$$

Linear viscosity, defined by the viscosity of Newton fluid [5], that is:

$$\sigma(t)=\eta\bar{\gamma}=k\frac{d\gamma}{dt} \tag{4}$$

With σ is the shear stress, $\bar{\gamma}$ is the rate of shear strain, η is the viscosity of the fluid, and t is time. Linear elasticity is defined by Hooke's Law [5], that is:

$$\sigma(t)=G\gamma=k\frac{d^0\gamma}{dt^0} \tag{5}$$

With σ is the shear stress, γ is a stretch and G is the modulus of elasticity or Young's modulus. Based on (4) and (5), for the material formed between liquid and solid, the following viscoelasticity formula is obtained:

$$\sigma(t)=k\frac{d^\alpha\gamma}{dt^\alpha} \tag{6}$$

Where $0<\alpha<1$. It means, if $\alpha=0$ then the material is a solid and if $\alpha=1$ then the material is a liquid. When the stress (force) is removed in the viscoelastic material, the material can not be directly returned to its original form, the Boltzmann principle of superposition is used to obtain the final voltage derived from all voltage additions [10], that is:

$$\begin{aligned}\sigma(t)&=k_1\frac{d^{\alpha_1}\gamma}{dt^{\alpha_1}}+k_2\frac{d^{\alpha_2}\gamma}{dt^{\alpha_2}}+\cdots+k_N\frac{d^{\alpha_N}\gamma}{dt^{\alpha_N}}\\&=\sum_{n=1}^N k_n\frac{d^{\alpha_n}\gamma}{dt^{\alpha_n}}=\sum_{n=1}^N k_n D^{\alpha_n}[\gamma(t)].\end{aligned} \tag{7}$$

Fourier transforms equation (7) according to (3), then:

$$\begin{aligned}F\{\sigma(t)\}&=F\left\{\sum_{n=1}^N k_n D^{\alpha_n}[\gamma(t)]\right\}\\ \sigma^*(\omega)&=\sum_{n=1}^N k_n F\left\{D^{\alpha_n}[\gamma(t)]\right\}=\sum_{n=1}^N k_n (i\omega)^{\alpha_n} F[\gamma(t)]\\&=\sum_{n=1}^N k_n (i\omega)^{\alpha_n}\gamma^*(\omega)\end{aligned} \tag{8}$$

Where $\sigma^*(\omega)$ and $\gamma^*(\omega)$ is the transformation of stress and strain, which is a representation of the complex stresses and strains. In addition, α_n and k_n are constants obtained from the experimental data, and ω is the angular frequency. Equation (8) is very difficult to practice because it contains too many constants, and then selected the first two forms of deformation, that is:

$$\sigma^*(\omega) = \left[k_1(i\omega)^{\alpha_1} + k_2(i\omega)^{\alpha_2}\right]\gamma^*(\omega) \tag{9}$$

With k1, k2, α_1, and α_2 is a parameter of the simplified model. Based on linear viscoelasticity, then the modulus of the complex is obtained:

$$G^*(\omega) = \frac{\sigma^*(\omega)}{\gamma^*(\omega)} = k_1(i\omega)^{\alpha_1} + k_2(i\omega)^{\alpha_2} \tag{10}$$

By delineating the equation (10), the real and imaginary part of the complex modulus (preliminary analysis): $i = e^{\frac{\pi}{2}i}$ and $e^{i\theta} = \cos\theta + i\sin\theta$ $e^{i\theta} = \cos\theta + i\sin\theta$).

$$\begin{aligned} G^*(\omega) &= k_1(i\omega)^{\alpha_1} + k_2(i\omega)^{\alpha_2} = k_1\omega^{\alpha_1}i^{\alpha_1} + k_2\omega^{\alpha_2}i^{\alpha_2} \\ &= k_1\omega^{\alpha_1}\left(e^{\frac{\pi}{2}i}\right)^{\alpha_1} + k_2\omega^{\alpha_2}\left(e^{\frac{\pi}{2}i}\right)^{\alpha_2} \\ &= k_1\omega^{\alpha_1}\left(\cos\left(\frac{\pi}{2}\alpha_1\right) + i\sin\left(\frac{\pi}{2}\alpha_1\right)\right) \\ &\quad + k_2\omega^{\alpha_2}\left(\cos\left(\frac{\pi}{2}\alpha_2\right) + i\sin\left(\frac{\pi}{2}\alpha_2\right)\right) \\ &= k_1\omega^{\alpha_1}\cos\left(\frac{\pi}{2}\alpha_1\right) + k_2\omega^{\alpha_2}\cos\left(\frac{\pi}{2}\alpha_2\right) \\ &\quad + i\left(k_1\omega^{\alpha_1}\sin\left(\frac{\pi}{2}\alpha_1\right) + k_2\omega^{\alpha_2}\sin\left(\frac{\pi}{2}\alpha_2\right)\right). \end{aligned}$$

From the previous equation, the complex modulus formula is obtained as follows:

$$G^*(\omega) = G'(\omega) + iG''(\omega) \tag{11}$$

Where $G'(\omega)$ is a storage modulus that shows the properties of elasticity and stored energy, and $G''(\omega)$ is the loss of modulus which indicates the nature of the viscosity and the energy released. Then the model for storage modulus and loss modulus, is given by:

$$G'(\omega) = k_1\omega^{\alpha_1}\cos\left(\frac{\pi}{2}\alpha_1\right) + k_2\omega^{\alpha_2}\cos\left(\frac{\pi}{2}\alpha_2\right), \tag{12}$$

$$G''(\omega) = k_1\omega^{\alpha_1}\sin\left(\frac{\pi}{2}\alpha_1\right) + k_2\omega^{\alpha_2}\sin\left(\frac{\pi}{2}\alpha_2\right). \tag{13}$$

3. Results

Base on Table 1 [5-15], the following models of viscoelasticity are storage and loss modulus xanthan gum solution for 0.5% concentration,

$$G'(\omega) = 15.08677792\omega^{0.3444} - 13.56167211\omega^{0.343},$$

$$G''(\omega) = 4.435826188\omega^{0.1622} - 4.312640582\omega^{0.176}.$$

For 1.0% concentration:

$$G'(\omega) = 35.46861744\omega^{0.1838} + 182.1021181\omega^{0.1842},$$

$$G''(\omega) = -59.18329232\omega^{0.1313} + 123.9497665\omega^{0.1318}.$$

For 2.0% concentration:

$$G'(\omega) = -313.0639973\omega^{0.2839} + 612.5702472\omega^{0.2304},$$

$$G''(\omega) = -20.21716628\omega^{0.138} + 104.2453882\omega^{0.1382}.$$

For 3.0% concentration:

$$G'(\omega) = -492.382097\omega^{0.2413} + 1060.673577\omega^{0.1905},$$

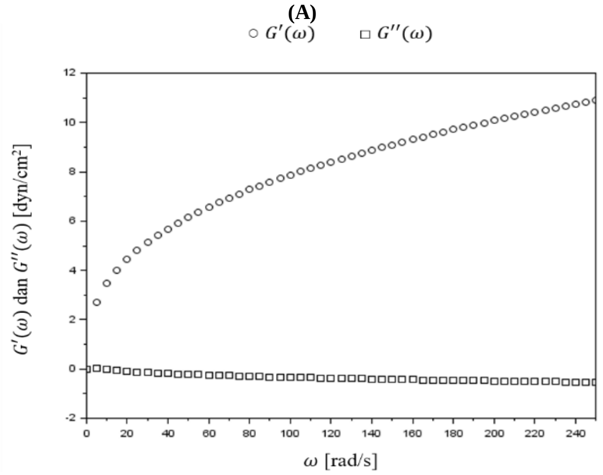
$$G''(\omega) = -17.2438974\omega^{0.1162} + 148.6330585\omega^{0.1169}.$$

For 4.0% concentration:

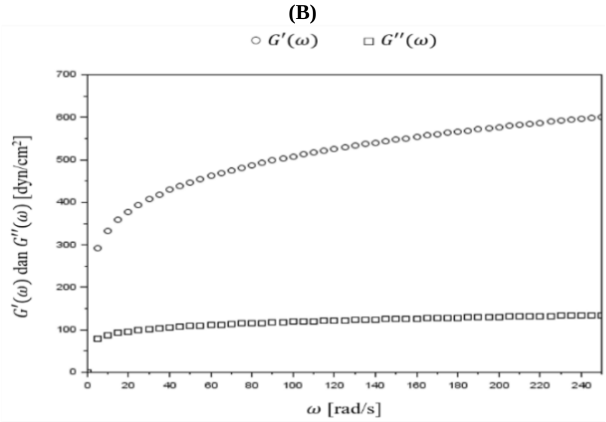
$$G'(\omega) = -380.8160379\omega^{0.2296} + 1207.484417\omega^{0.1646},$$

$$G''(\omega) = -700.544122\omega^{0.1123} + 883.5770677\omega^{0.1123}.$$

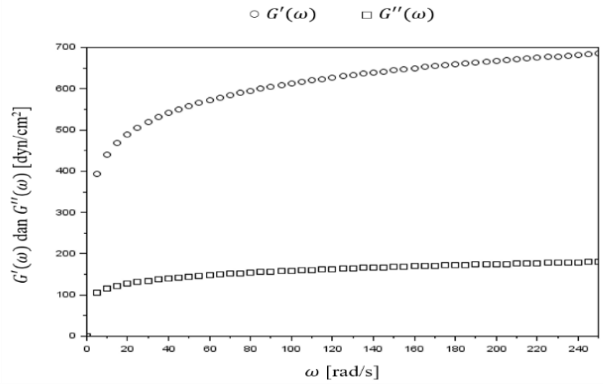
(A)



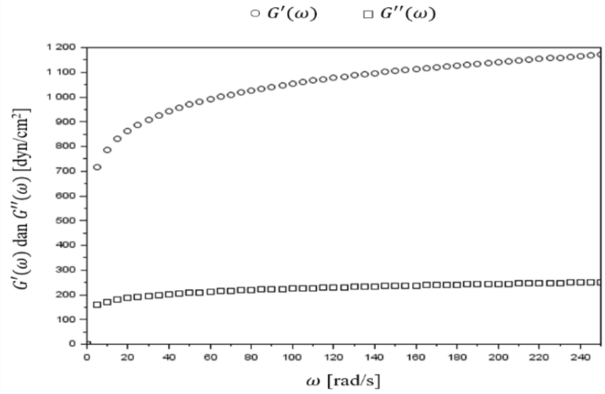
(B)



(C)



(D)



(E)

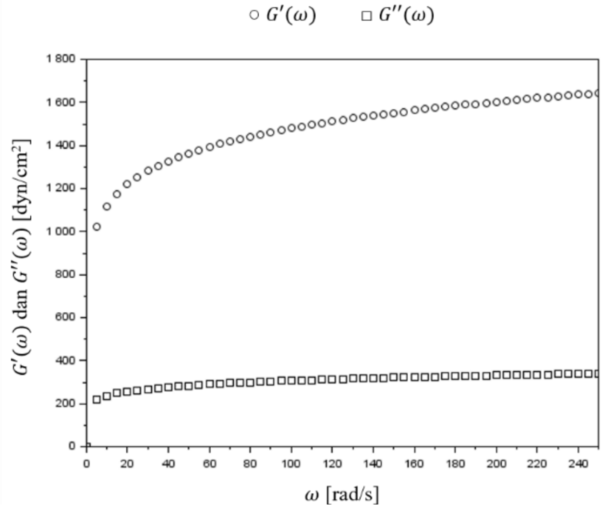
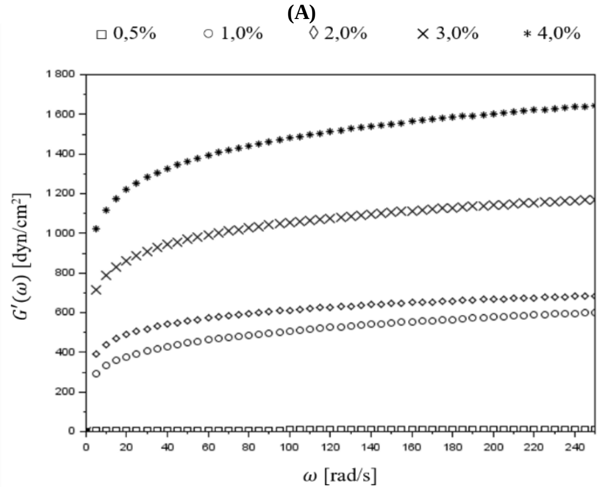


Fig. 1: Storage And Loss Modulus In Xanthan Gum Solution with Concentration: (A) 0.5%; (B) 1.0%; (C) 2.0%; (D) 3.0%; and (E) 4.0%.

(A)



(B)

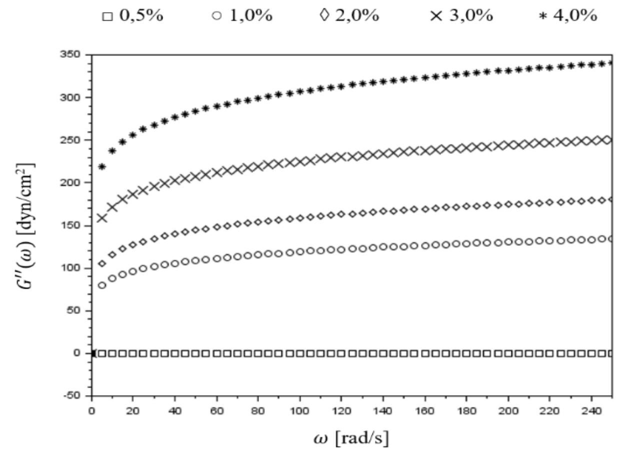


Fig. 2: Xanthan Gum Solution with Concentrations of 0.5%, 1.0%, 2.0%, 3.0%, and 4.0%: (A) Modulus Storage (B) Loss of Modulus.

4. Discussion

Based on Figure 1 (a), storage and loss modulus in xanthan gum solution with 0.5% concentration, it can be seen that the storage graph of monotonous modulus increases, while the monotonous modulus loss decreases. Based on Figure 1 (b) ~ (e), it can be seen that both storage graphs and monotonous modulus losses rise. In addition, for every angular frequency, the modulus storage value is always greater than its modulus loss value. This suggests that the elastic properties of the xanthan gum solution are more dominant than their viscous (viscous properties) for all concentrations.
Based on Figure 2 (a), the storage graph of the xanthan gum solution modulus for all concentrations, the more concentrated the xanthan gum solution, the more its modulus storage value. This is the same for the graph of modulus loss for all concentrations, based on Figure 2 (b), the increasing concentration of xanthan gum solution is also increasing as well as its modulus loss value.
Fractional derivative models with Riemann-Liouville approach had good ability to investigate the viscoelastic behavior of xanthan gum solution with concentrations of 0.5%, 1.0%, 2.0%, 3.0%, and 4.0%. However, this model may be classified as semi-empirical relationships because there was no real physical significance for the model parameters. In addition, this model can only be used specifically for viscoelasticity in xanthan gum solution.
Based on the viscoelasticity of xanthan gum model that shown by equations (12) and (13), storage and loss modulus respectively, then will be analyzed using simulation data. simulation data fr storage modulus is determined randomly for value k1 at 0.5% concentration which is 22.1, while k2 is obetained from the sum of the difference between k1 and k2 in experimental data. Then k1 for 1.0%, 2.0%, 3.0%, and 4.0% obtained by adding the difference of k1 with the previous concentration. The same way is also done to obtained loss modulus simulation data by randomly determining the value of k1 0.5% concentration which is 15.3, but the value of both α_1 and α_2 is not given change. So the simulation data is obtained as follows.

Table 2: Simulation Data

Concentration	G’ and G”	k_1	k_2	α_1	α_2
0.5%	G’	22.1	-11.3	0.3444	0.3430
	G”	15.3	-18.1	0.1622	0.1760
1.0%	G’	4.15	194.5	0.1838	0.1842
	G”	-291.3	600.7	0.1313	0.1318
2.0%	G’	-342.5	659.5	0.2839	0.2304
	G”	-96.3	481.7	0.1380	0.1382
3.0%	G’	-525.5	1114.5	0.2413	0.1905
	G”	-97.3	811.7	0.1162	0.1169
4.0%	G’	-402.5	1253.5	0.2296	0.1646
	G”	-3994.3	5032.7	0.1123	0.1123

Then the simulation data from Table 2 is substututed into equations (12) and (13), so fo 1.0% concentration the graph has shown in Figure 3.

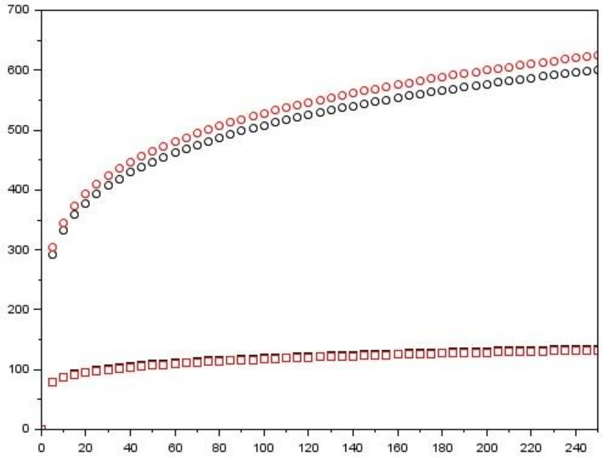


Fig. 3: Simulation Graphic.

Figure 3 shows that the graph of simulation data approaches or has same caracteristic as the graph of experimental data.

5. Conclusion

Based on the results of this research, one application of fractional derivatives is to determine the viscoelasticity model in xanthan gum. From the analysis of viscoelasticity model, it can be concluded that both storage and loss modulus for all concentrations increase with increasing angular frequency, except in loss modulus with 0.5% concentration. In addition, all modulus storage values outweigh the modulus loss for each angular frequency. This suggests that the elastic properties of the xanthan gum solution are more dominant than their viscous/viscous properties for all concentrations. In addition, there is an increase in modulus storage along with the increased concentration of xanthan gum solution; this applies also to the loss of modulus. That is, the xanthan gum solution is getting more elastic as the concentration of the solution increases. It can be seen that the greater the concentration of the solution the slower the relaxation mechanism. The fractional derivative model Riemann-Liouville has a good ability to investigate the viscoelastic behavior of all concentrations of xanthan gum solution. However, this model may be classified as semi-empirical relationships because there is no real physical significance for model parameters. In addition, this model can only be used specifically for viscoelasticity in xanthan gum solution. So, it is expected that in later research this viscoelastic model can be applied for materials besides xanthan gum; and it would be better if a simulation with more data, including trying to change the value of α_1 and α_2.

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