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RHEOLOGICAL MODEL OF VISCOUS INCOMPRESSIBLE FLUID WITH DISPERSE FILLERS

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ABSTRACT

The article is devoted to a relevant problem of the production of composite structures, namely finding ways to reduce the duration of fibrous filler impregnation in the technological process of forming, one of which is increasing the shear rate when moving the binder in the intrafibrous space of the construction wall.

The aim of this work is to study mechanisms of the shear rate influence on the binder viscosity. During the study the methods of mathematical modeling of viscous fluid shearing movement with the subsequent experimental adjustment of models were used.

By the example of polymethylphenylsiloxane the nature of change of viscosity dependence on shear rate at different content of solid particles of filler in viscous liquid was analyzed. The obtained mathematical models explain the effects of the formation of clusters under the close interaction forces between the liquid molecules and filler particles on the rheological properties of viscous fluids. Cluster size limits the thermal oscillation of cluster elements and their rotation when shift rates are available.

It was found that power dependence between viscosity and shear rate provides minimum energy consumption. Experimental data processing showed that the power exponent value increases as the volume fraction of the filler changes. Analysis of experimental data has shown that the mathematical expression for viscosity description has invariant and variant parts. The first one characterizes the reversibility of viscosity changes with shear rate increasing and decreasing. The second part, depending on the derivative sign of the time shear rate will have a different value.

Key words: Binder, Rheological properties, Disperse filler, Physical chemistry

1. INTRODUCTION

In the technological processes of production of composite structures the essential factor for providing the quality and required technical and economic indices is viscosity of binders. In particular in the works [1-5] the issues of organization of processes of winding and contact molding considering the viscosity influence are discussed. Ways of its reducing are sought [6-9]. A large number of applications is known, where viscosity is an important factor in the thermal coating impregnation technologies of space ships of an ablative type [10-16] and the technologies of manufacturing of porous thermal coatings [17, 18].

Rheological properties of viscous liquids are a manifestation of hidden processes of fluid restructuring when external impact conditions change. An essential quantity of works is dedicated to its studying [19-21]; the large volume of experimental data is accumulated in them. [19-21]. In particular, the role of fillers in ensuring the viscosity of binders was reviewed. From the standpoint of theoretical foundations of physical chemistry the models of formation of cluster structures were offered, whose properties depend on the type and amount of filler.

Along with this the models proposed in the works [22-24] are qualitative in nature and does not create conditions for mathematical modelling of processes, because the questions of the restructuring description is difficult to formalize.

Therefore, in our opinion, the wording of approaches is interesting, which, based on well-known mechanical (hydromechanical) views, will allow approaching to the mathematical formalization of the experimental data. The present article is devoted to this.

2. MATERIALS & EXPERIMENTAL PROCEDURES

We will use the experimental data [22, 24] on rheology of polymethylphenylsiloxane of the brand SKTNF with elastomeric additives (fig. 1), which solidification is possible at room temperature by means of aminnym hardener.
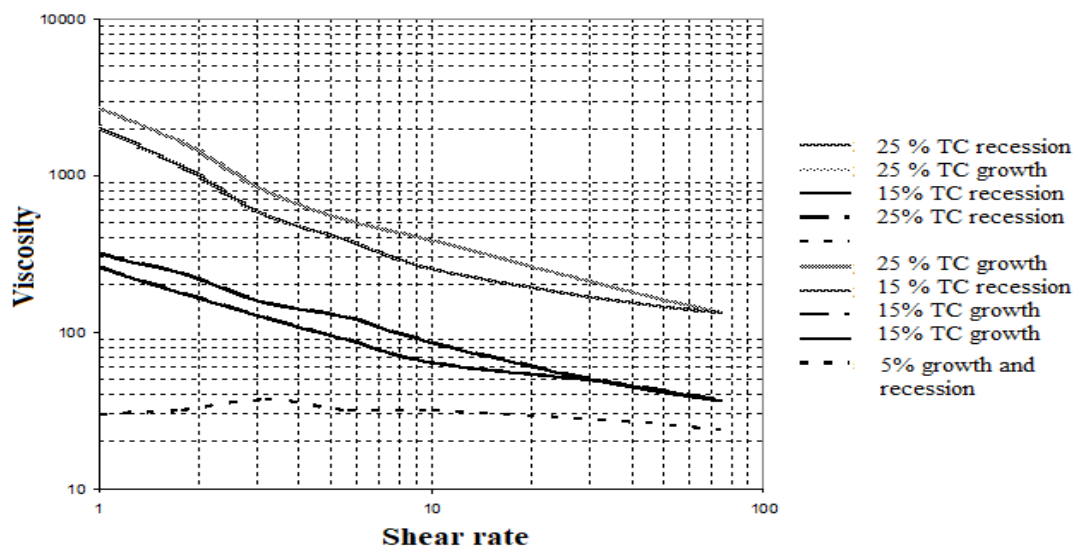

Figure 1. Dependencies of polymethylphenylsiloxane viscosity of the brand SKTNF at different mass fraction of technical carbon additive (TC) at its content (5, 15, 25%) obtained with the increase (growth) and decrease (recession) of the shear rate.

As filler the technical carbon (TC) with liophobic-lyophilic surface and iron-carbon powder with lyophilic -liophobic surface was used.

3. RESULTS AND DISCUSSION

Experimental data show that in the plane viscosity-shear rate each TC mass proportion corresponds to its own curve. When you increase the shear rate the general tendency is viscosity decrease. Along with TC content increasing the viscosity also increases.

An interesting phenomenon consists in the disparity of dependencies between viscosity and shear rate with the increasing and decreasing of the shear rate. On the curves there is an inversion point to the right of which the value of viscosity is the same at the growth and recession of shear rate. For TC mass fraction 15 and 25% to the left of an inversion point the values of viscosity at the growth and recession of shear rate varies. This phenomenon is called hysteresis, and depending on the type of filler there may be the phenomena of normal (when the viscosity at the shear rate increase prevail the viscosity at the reduction of this setting) and abnormal hysteresis (when the viscosity at the shear rate increase is less than the viscosity at the recession of this parameter).

From the standpoint of physics foundations there are forces of close interaction between the particles of filler and viscous liquid that cause their agglomeration with the formation of clusters. However, as well as in metals with a polycrystalline structure the formation of clusters occurs around the formation centers simultaneously. And on the borders of clusters their structure is misoriented in space and the connections between clusters are weakened.

Formal description of the mentioned phenomenon can be carried out in accordance with the methodology described in the work [25], assuming that the action of attractive force between the volume ϑ_1 and the unit volume ϑ^* obeys the power dependence

$$F_{pr} = \gamma_{pr} \frac{\vartheta_1 \vartheta^*}{r^n} \tag{1}$$

where r - distance between volumes γ_{pr} - proportionality constant.

In accordance with this law, the attractive force change of the unit volume ϑ^* on the cluster surface F_{pr}^s and the change in the work of separation A_{pr}^s of the unit volume ϑ^* from the cluster surface are described by the ratios

$$F_{pr}^s = \frac{4}{3}\pi\gamma_{pr}\frac{\vartheta^*}{r_{agl}^{n-3}}, \qquad A_{pr}^s = \frac{4}{3}\pi\frac{\gamma_{pr}\vartheta^*}{(n-1)r_{agl}^{n-4}} \tag{2}$$

Cluster size is limited by the energy of thermal vibrations of binder molecules that you can adopt by the proportional internal energy in the volume ϑ^*, i.e.

$$E_{ost} = K_{ost}\rho C_V \vartheta^* \Delta T \tag{3}$$

where ρ, C_V, ΔT -density, heat capacity, temperature of binder.

Equating E_{ost} with A_{pr}^s, we obtain at $n \geq 5$ the cluster growth restriction condition

$$r_{kl} \leq \left[\frac{4\pi}{3(n-1)}\frac{\gamma_{pr}}{K_{ost}\rho C_V \Delta T}\right]^{\frac{1}{n-4}} \tag{4}$$

From the hydromechanical positions the shear rate is one of the components of rotor of mass velocity vector in the viscous liquid stream. So one can consider that it is the shear deformations occur at the progressive and rotational motion of clusters.

Rotation of the cluster and the related process of sedimentation of binder components can be another restrictive factor. At the cylinder rotation with diameter r_{kl} the distribution of stresses in his body is described by the differential equation

$$\frac{d\sigma_m}{dr} + \frac{\sigma_m - \sigma_\theta}{r} - \rho\omega^2 r = 0 \tag{5}$$

where σ_m, σ_θ - meridional and circumferential stresses, ρ - liquid density, ω - the angular velocity of sinter rotation, r - sinter layer coordinate from its axis.

For liquids the equity of stress components is usually taken $\sigma_m = \sigma_\theta = \sigma_x = -p$. In this case, the solution to the equation takes the form

$$p = -\frac{1}{2}\rho\omega^2 r^2 \tag{6}$$

Maximum negative pressure will correspond to the sinter periphery

$$p\frac{1}{2}^{2^2}_{kl_{max}} \tag{7}$$

The appearance of negative pressure corresponds to the increase of the sinter volume

$$p = -\frac{4}{3}\pi\gamma_{pr}\frac{(n-3)}{r_{kl}^{n-3}}\frac{\Delta r}{r_{kl}} = -\frac{4}{9}\pi\gamma_{pr}\frac{(n-3)}{r_{kl}^{n-3}}\frac{\Delta\vartheta}{\vartheta} \tag{8}$$

Hence

$$\frac{\Delta\vartheta}{\vartheta} = -\frac{9r_{kl}^{n-3}p}{4\pi\gamma_{pr}(n-3)} \tag{9}$$

Work of volume unit deformation of sinter

$$A^0 = -\int_0^{\Delta\vartheta} p\, d\,\Delta\vartheta = \frac{2}{9}\pi\gamma_{pr}\frac{(n-3)}{r_{kl}^{n-3}}\frac{\Delta\vartheta^2}{\vartheta} = -\frac{1}{2}p\Delta\vartheta = \frac{p^2}{2K} \tag{10}$$

Where $K = \frac{4\pi\gamma_{pr}(n-3)}{9r_{kl}^{n-3}\vartheta^*}$

If we present the condition of sinter destruction at rotation in the form of $A^0 = A_{pr}^s$, we'll get another sinter size restriction.

$$r_{kl} \leq \left[\frac{n-3}{n-1}\right]^{\frac{1}{2n-3}}\left[\frac{0,253\pi\gamma_{pr}}{\rho\omega^2}\right]^{\frac{1}{n-1,5}} \tag{11}$$

The work of internal friction forces defines three processes occurring in the viscous fluid: the formation of vortices of size $a \sim r_{kl}$, volumetric expansion of clusters, which takes work A_{str}, as well as the work of frictional forces on the borders of vortices. At that the parameters of these processes must comply with the minimum power intensity of the process.

If the cell size with the vortex is a, and the formation of the unit of division plane between vortices takes specific work A^*, in the region of the internal friction impact of size l there will be $N^2 = \left(\frac{l}{a}\right)^2$ of division planes between vortices, and the work of the formation of division planes

$$A_{for} = A^* l \left(\frac{l}{a}\right)^2 \tag{12}$$

When the velocity gradient $\xi = \frac{v}{l}$ and the size of the vortex cell a the rotation velocity is

$\omega = \xi\frac{a}{2}\frac{2}{a} = \xi$. Sinter rotation causes negative pressures

$$p = -\frac{1}{2}\rho\omega^2 r^2 \tag{13}$$

The potential energy of the sinter expansion is equal to

$$A_{str} = -\int_0^{\Delta\vartheta} A^0 \, \Delta\vartheta = \frac{\rho^2}{24\pi^2 K}\omega^4 a^6 \tag{14}$$

Power intensity of the process will be characterized by the parameter

$$A_\Sigma = A_{for} + A_{str}\left(\frac{l}{a}\right)^2 \tag{15}$$

The minimum power intensity of the process must comply with the condition $\frac{dA_\Sigma}{da} = 0$. Hence one can get an estimate of the vortex size

$$a = \left(\frac{2\pi\sqrt{3A^* l K}}{\rho\xi^2}\right)^{\frac{1}{3}} \sim r_{kl} \tag{16}$$

The minimum power intensity of the process will be connected with the velocity gradient of the viscous fluid flow by the power dependence

$$A_\Sigma = \frac{l^2}{2}\left(\frac{3A^* l\rho\xi^2}{2\pi\sqrt{K}}\right)^{\frac{2}{3}} = \frac{3A^* l^3}{2a^2} \sim \frac{3A^* l^3}{2r_{kl}{}^2} \tag{17}$$

Alternatively

$$A_\Sigma \sim \tau\xi = \mu\xi^2 \sim \frac{l^2}{2}\left(\frac{3A^* l\rho\xi^2}{2\pi\sqrt{K}}\right)^{\frac{2}{3}} \tag{18}$$

As a result of conversions for the value μ the dependency occurs

$$\frac{\mu}{\mu_0} = \left(\frac{\xi_0}{\xi}\right)^{\frac{2}{3}} \tag{19}$$

where ξ_0, μ_0 - minimum shear rate in the experiments and viscosity value corresponding to the minimum percentage of carbon.

Processing the experimental data, we will take a generalized form of power dependence

$$\bar{\mu} = \frac{A*}{\bar{\xi}m} \tag{20}$$

where $\bar{\mu} = \frac{\mu}{\mu_0}$- relative value of viscosity, $\bar{\xi} = \frac{\xi}{\xi_0}$ - relative value of shear rate.

The required dependency in the form of graphs in the plane $l\bar{g}\,\mu \quad - \quad l\bar{g}\,\xi$ is presented on the Fig.2

Presenting the dependency (20) in the form

$$l\bar{g}\,\mu = A - m\,l\bar{g}\,\xi \tag{21}$$

and processing the experimental data, we will get for the approximating equations (20) the dependency of parameters $A = \lg A^*$ and m as function from the proportion of technical carbon added to resin.

$$A = \begin{cases} 0{,}0552q\text{-}0{,}0009q^2 & \text{for the growth stage } \bar{\xi} \\ 0{,}0011 + 0{,}0847q & \text{for the recession stage } \bar{\xi} \end{cases} \tag{22}$$

For the growth stage

For the recession stage

$$m = \begin{cases} 0{,}45\text{-}0{,}0059q + 0{,}0006q^2 & \text{for the growth stage } \bar{\bar{\xi}} \\ 0{,}45 + 0{,}0084q + 0{,}001q^2 & for\ the\ recession\ stage\ \bar{\bar{\xi}} \end{cases} \qquad . \tag{23}$$
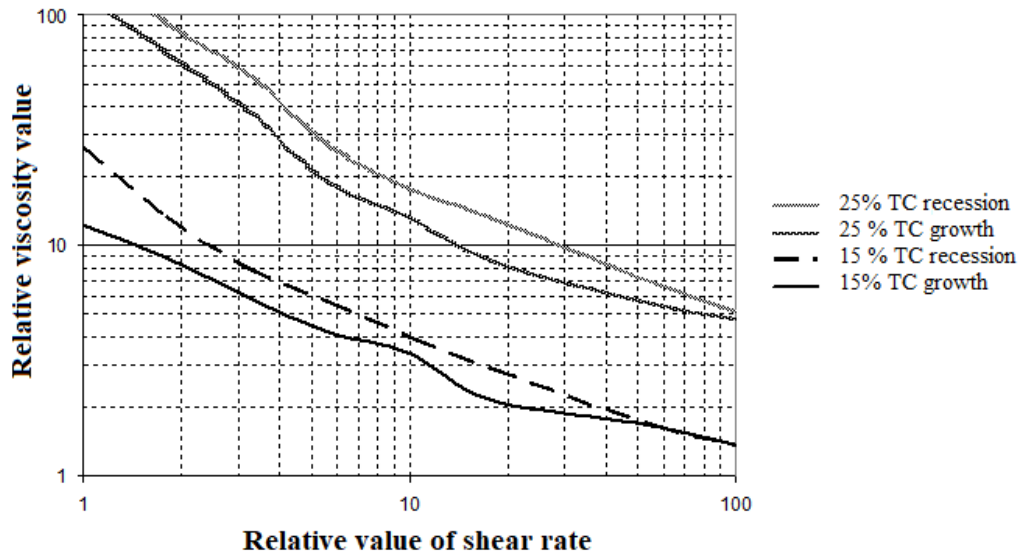
Figure 2. Dependence of the relative viscosity value $\bar{\mu}$ from the relative value of shear rate $\bar{\bar{\xi}}$ at different TC percentage at the stages of shear rate growth and recession.

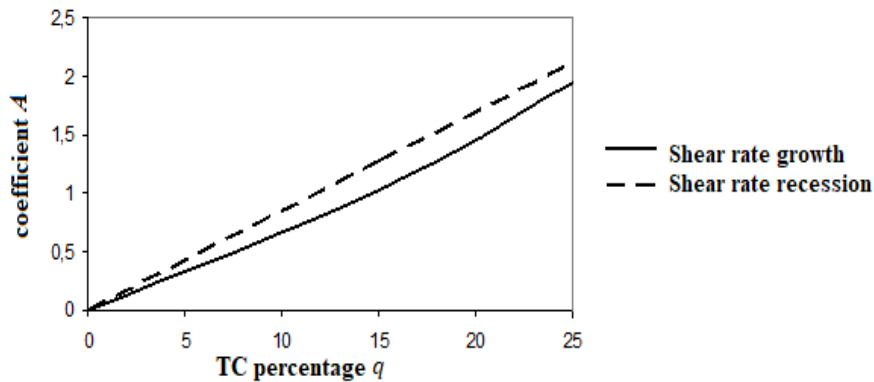The nature of these dependencies can be observed in Figures 3, 4.

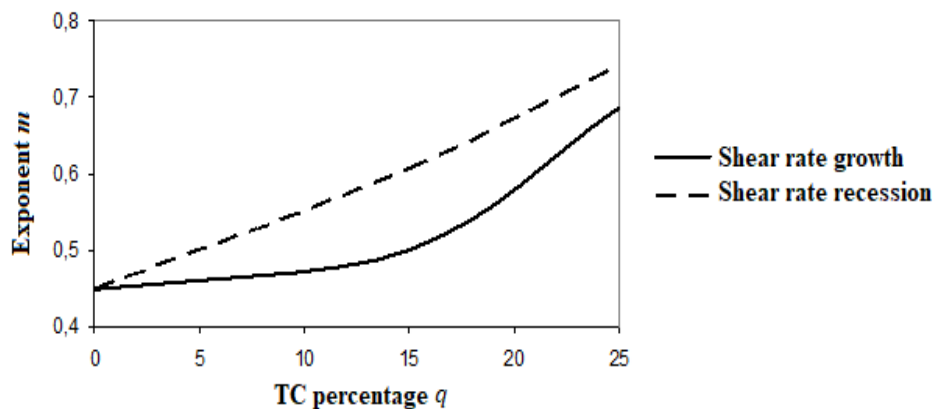**Figure 3.** Dependence of the values of coefficient A from TC percentage q

Figure 4. Dependence of the values of Exponent m from TC percentage q

The interesting object of study is a hysteresis phenomenon in the neighborhood of inversion points that occurs when you change the derivative sign $\frac{d\xi}{dt}$. A graphical representation of this phenomenon is given in Figure 5 in the coordinates "viscosity-shear rate", whereby the viscosity coefficient has invariant and variant parts.
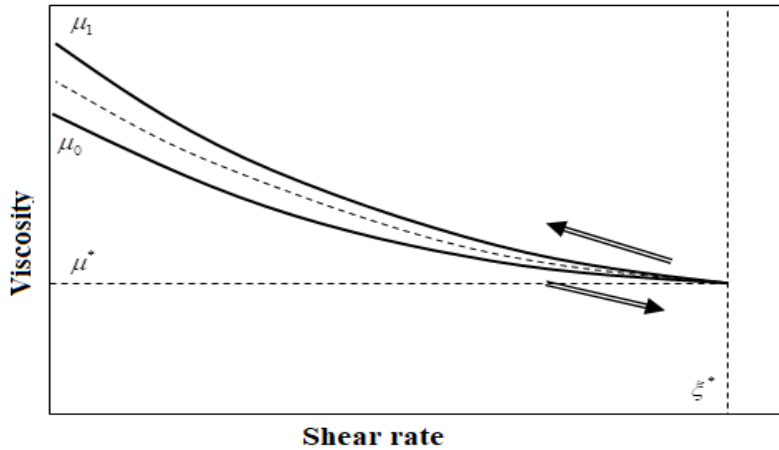

Figure 5. Graphic representation of hysteresis phenomena of dynamic viscosity at the inversion point ξ^*, μ^* at the shear rate growth and recession.

The invariant part corresponds to the dotted line, determined by the shear rate ξ and characterizes the reversibility of the process of change μ at the growth and recession ξ. The variant part is determined by the value $\dot{\xi} = \frac{d\xi}{dt}$ and will be of different value μ depending on the sign $\dot{\xi}$. Functional connection of viscosity and shear rate with consideration of the increase and decrease of $\dot{\xi}$ in accordance with the direction of the arrows in Fig. 5. is characterized by two solid lines. At the increase of ξ ($\dot{\xi} \geq 0$) the viscosity decreases from the value μ_0. After reaching the inversion point ξ^*, μ^* at the further decrease of ξ ($\dot{\xi} \leq 0$) the viscosity increases to the value μ_1. In the graph the value of the variant part corresponds to the deviation of points of the solid lines from the dotted line.

Formally, the dynamic viscosity coefficient can be represented as a function of the shear stresses and the velocity gradient

$$\mu = \mu\left(\xi, \dot{\xi}\right) \tag{24}$$

that will characterize the ever-changing structure of viscous fluid, where $\xi = \vec{\nabla}v$ - velocity gradient of the flow v, $\vec{\nabla}$ - Hamilton operator, $\dot{\xi} = \frac{d\xi}{dt}$ - the rate of the parameter change ξ with time t

For the mathematical description of hysteresis nearby the inversion point we will accept the equation of bunch of straight lines in the logarithmic coordinates

$$\begin{cases} lg\,\dfrac{\bar{\mu}}{\bar{\mu}^*} = -K\,lg\,\dfrac{\bar{\dot{\xi}}}{\bar{\dot{\xi}}^*} \\ K = \dfrac{m\uparrow + m\downarrow}{2} + \dfrac{\dot{\xi}}{|\dot{\xi}|}\dfrac{m\uparrow - m\downarrow}{2} \end{cases} \tag{25}$$

Where $lg\,\bar{\dot{\xi}}^* = \frac{A\downarrow - A\uparrow}{m\downarrow - m\uparrow}, lg\,\bar{\mu}^* = A\uparrow - m\uparrow\frac{A\downarrow - A\uparrow}{m\downarrow - m\uparrow}$, the symbols $\uparrow, \downarrow$ indicate the growth ($\dot{\xi} \geq 0$) or the decline ($\dot{\xi} \leq 0$) of the shear rate ξ, $A\uparrow, m\uparrow$- coefficient A and index m at the shear rate growth, $A\downarrow, m\downarrow$- coefficient A and index m at the shear rate decline.

4. CONCLUSIONS

- The process of forming clusters under the forces of close interaction between the liquid molecules and filler particles affects the rheological properties of viscous fluids.

- Cluster size is limited by the heat fluctuations of elements of clusters and their rotation when the shear rates are available.

- Theoretical analysis of energy consumption during viscous fluid flow revealed that power dependence between viscosity and shear rate provides minimum energy consumption.

- Processing of experimental data showed that the index value is hardly changed at close range of the theoretically found value. Deviations from the theoretically found value amount to.

- Analysis of the experimental data has shown that the mathematical expression to describe the viscosity has invariant and variant parts. The first one describes the reversibility of viscosity changes at the shear rate increasing and decreasing. The second part depending on the sign of the derivative of the time shear rate will have a different value.

- Of interest is the application of the proposed approach in the analysis of experimental data for other kinds of organic and inorganic binders.

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