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A ZERO-PARAMETER CONSTITUTIVE RELATION FOR PURE SHEAR
VISCOELASTICITY

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ABSTRACT

Based on the Cox-Merz rule and Eyring's expression for the nonlinear shear viscosity, a Wagner type constitutive relation with no adjustable parameters is proposed for pure shear viscoelasticity. The predictions for a number of non-steady shear flows are worked out analytically. It is shown that most features of shear viscoelasticity are reproduced by the model.

1. INTRODUCTION

After several years of research a number of useful constitutive relations are now available [1]. In order to reproduce experiment accurately these relations all contain a number of fitting parameters. In this paper the following question is asked: What is the simplest possible constitutive relation which still reproduces important features of viscoelasticity? To simplify the discussion only pure shear viscoelasticity is considered, and normal stresses are ignored all together. Starting from the Cox-Merz rule, a simple Wagner type constitutive relation with no adjustable parameters is arrived at. The nonlinear steady state shear viscosity is, by construction, close to that predicted by Eyring's phenomenological theory of liquid flow [2]. Various non-steady shear flows are then considered and worked out analytically. Because there are no fitting parameters in the proposed model, no detailed comparison to experiment is made. However, it is shown that the constitutive relation reproduces most qualitative features of shear viscoelasticity.

2. THE MODEL

The well-known Cox-Merz rule [3] states that

$$\eta(\dot{\gamma}) = \left| \eta_0(\omega) \right| \Big|_{\omega = \dot{\gamma}} \quad (1)$$

where $\eta(\dot{\gamma})$ is the nonlinear shear viscosity as function of shear rate and $\eta_0(\omega)$ is the frequency-dependent viscosity in the linear response regime. The Cox-Merz rule is obeyed surprisingly well for most polymeric liquids, while exceptions are sometimes observed for inhomogeneous systems. The quantity $\eta_0(\omega)$ is obtained [1] from the equation

$$\eta_0(\omega) = \int_0^{\infty} dt' G(t') e^{-i\omega t'} \quad (2)$$

where $G(t')$ is the relaxation modulus. By definition, $G(t')$ determines the stress τ in the linear limit from the shear rate history by means of

$$\tau(t) = \int_0^{\infty} dt' G(t') \dot{\gamma}(t-t'). \quad (3)$$

From Eqs. (1) and (2) one expects the Cox-Merz rule to be satisfied if

$$\eta(\dot{\gamma}) = \int_0^{\infty} dt' G(t') e^{-\dot{\gamma} t'} \quad (\dot{\gamma} > 0). \quad (4)$$

A straightforward generalization of Eq. (3) to include Eq. (4) for

the stationary case is the following constitutive relation

$$\tau(t) = \int_0^{\infty} dt' G(t') \dot{\gamma}(t-t') \exp\left\{-\int_{t-t'}^t |\dot{\gamma}(t'')| dt''\right\} \quad (5)$$

Equation (5) is similar to Wagner's constitutive relation [1,4].

The difference is that, in the "linear" part of the relation,

γ in Wagner's model is here replaced by $\dot{\gamma}$. More importantly, the "damping function" is here $\exp\left\{-\int_{t-t'}^t |\dot{\gamma}| \right\}$

instead of Wagner's $\exp\left\{-\int_{t-t'}^t \dot{\gamma} \right\}$. The present choice of

damping function is suggested because this damping function sums over all shear displacement taking place between time $t-t'$ and

t .

Next, a specific form of $G(t')$ is chosen, namely $G(t') = E_1(t')$ where $E_1(t')$ is the exponential integral [5]

$$E_1(t') = \int_{t'}^{\infty} e^{-u} \frac{du}{u} \quad (6)$$

For convenience we here and henceforth work with dimensionless time, stress and viscosity, the latter quantity normalized so that $\eta_0(\omega=0) = 1$. The final constitutive relation is

$$\tau(t) = \int_0^{\infty} dt' E_1(t') \dot{\gamma}(t-t') \exp\left\{-\int_{t-t'}^t |\dot{\gamma}(t'')| dt''\right\} \quad (7)$$

The use of $E_1(t')$ as the relaxation modulus is motivated by the fact that this choice leads to a nonlinear viscosity which is close to that predicted by Eyring's phenomenological theory of liquid flow [2] which fits many experiments:

$$\eta(\dot{\gamma}) = \frac{\sinh^{-1}(\dot{\gamma})}{\dot{\gamma}} \quad (8)$$

To see this, note that the Laplace transform of E_1 is [5]

$$\tilde{E}_1(s) = \frac{\ln(1+s)}{s} \quad (9)$$

so the nonlinear viscosity is given by

$$\eta(\dot{\gamma}) = \frac{\ln(1+\dot{\gamma})}{\dot{\gamma}} \quad (10)$$

From the identity $\sinh^{-1}(x) = \ln(x + \sqrt{1+x^2})$ it follows that Eyring's nonlinear shear viscosity is close to that predicted in Eq. (10). This is illustrated in Fig. 1 which also shows that the Cox-Merz rule is obeyed by the constitutive relation, as expected. This observation is based on the fact that the frequency-dependent linear viscosity is given by

$$\eta_0(\omega) = \int_0^{\infty} dt' E_1(t') e^{-i\omega t'} = \frac{\ln(1+i\omega)}{i\omega} \quad (11)$$

which implies for the real part and for the (negative) imaginary part

$$\begin{aligned} \eta'(\omega) &= \text{Arctan}(\omega) / \omega \\ \eta''(\omega) &= \ln \sqrt{1+\omega^2} / \omega \end{aligned} \quad (12)$$

We now proceed to calculate the time-dependent nonlinear response in various situations, following Chap. 3.4 in Ref. 1.

Consider first the stress growth upon inception of a steady shear flow, i. e. , the case when the shear rate is given by

$$\dot{\gamma}(t) = \begin{cases} 0, & t < 0 \\ \dot{\gamma}_0, & t > 0 \end{cases} \quad (13)$$

In this case Eq. (7) implies for the stress τ^+ :

$$\tau^+(t) = \dot{\gamma}_0 \int_0^t dt' E_1(t') e^{-\dot{\gamma}_0 t'} \quad (14)$$

or, for the quantity $\eta^+(t, \dot{\gamma}_0) = \tau^+(t) / \dot{\gamma}_0$,

$$\eta^+(t, \dot{\gamma}_0) = \int_0^t dt' E_1(t') e^{-\dot{\gamma}_0 t'} \quad (15)$$

After a partial integration Eq. (15) reduces to

$$\eta^+(t, \dot{\gamma}_0) = \left[E_1((1+\dot{\gamma}_0)t) - E_1(t) e^{-\dot{\gamma}_0 t} + \ln(1+\dot{\gamma}_0) \right] / \dot{\gamma}_0 \quad (16)$$

where use has been made of the fact that $E_1(t)$ varies as $\ln(t)$ for $t \rightarrow 0$. In Fig. 2 $\eta^+(t, \dot{\gamma}_0)$ is plotted in a logarithmic plot for different values of $\dot{\gamma}_0$. The figure shows that η^+ is always monotonously increasing. This is not quite like in experiment where there is usually a characteristic "overshoot" of η^+ as function of time before the steady state value is reached [1].

Consider now stress relaxation after cessation of a steady shear flow, i. e. , when

$$\dot{\gamma}(t) = \begin{cases} \dot{\gamma}_0, & t < 0 \\ 0, & t > 0 \end{cases} \quad (17)$$

Then Eq. (7) implies for the stress $\bar{\tau}$:

$$\bar{\tau}(t) = \dot{\gamma}_0 \int_t^{\infty} dt' E_1(t') e^{-\dot{\gamma}_0(t'-t)} \quad (18)$$

Equations (10), (15) and (18) imply

$$\eta^+(t, \dot{\gamma}_0) + e^{-\dot{\gamma}_0 t} \eta^-(t, \dot{\gamma}_0) = \frac{\ln(1 + \dot{\gamma}_0)}{\dot{\gamma}_0} \quad (19)$$

where $\eta^-(t, \dot{\gamma}_0) = \bar{\tau}(t) / \dot{\gamma}_0$. By means of Eq. (16) we thus find

$$\eta^-(t, \dot{\gamma}_0) = \left[E_1(t) - E_1((1 + \dot{\gamma}_0)t) e^{\dot{\gamma}_0 t} \right] / \dot{\gamma}_0 \quad (20)$$

Fig. 3 shows η^- for various values of $\dot{\gamma}_0$. As in experiment one finds that $\eta^-(t, \dot{\gamma}_0)$ is a monotonously decreasing function of time for all $\dot{\gamma}_0$, and that η^- reaches zero faster the larger $\dot{\gamma}_0$ is.

We now turn to the calculation of stress relaxation after a sudden shearing displacement γ_0 . The shear rate is given by $\dot{\gamma}(t) = \dot{\gamma}_0 \delta(t)$. Substituted into Eq. (7) this gives

$$\bar{\tau}(t) = (1 - e^{-\dot{\gamma}_0 t}) E_1(t) \quad (21)$$

which is easily shown by rewriting Eq. (7) as

$$\bar{\tau}(t) = \int_0^{\infty} dt' E_1(t') \left[-\frac{d}{dt'} \right] e^{-\int_{t-t'}^t \dot{\gamma}} \quad (22)$$

valid whenever $\dot{\gamma} \geq 0$. For the relaxation modulus $G(t, \dot{\gamma}_0) = \bar{\tau}(t) / \dot{\gamma}_0$, one thus finds

$$G(t, \gamma_0) = E_1(t) (1 - e^{-\gamma_0}) / \gamma_0 \quad (23)$$

For $\gamma_0 \rightarrow 0$ $G(t, \gamma_0)$ reduces to the linear response relaxation modulus $E_1(t)$. Equation (23) shows that $G(t, \gamma_0)$ factorizes into a function of time times a function of γ_0 , as expected for any Wagner type model [1,6].

Next we consider the calculation of the nonlinear creep compliance $J(t, \bar{\tau}_0)$, defined as $\gamma(t)/\bar{\tau}_0$, where $\gamma(t)$ is the total shear displacement in time t when a constant stress $\bar{\tau}_0$ is applied at $t=0$. The calculation of J from a constitutive relation is complicated by the fact that $\gamma(t)$ is only given indirectly. For the present constitutive relation, however, $\gamma(t)$ may be found analytically in the following way. First, Eq. (7) is rewritten for the case under consideration as

$$\bar{\tau}_0 e^{\gamma(t)} = \int_0^t dt' E_1(t') \dot{\gamma}(t-t') e^{\gamma(t-t')} \quad (24)$$

Equation (24) is linear in the variable $C(t) = \exp(\gamma(t))$:

$$\bar{\tau}_0 C(t) = \int_0^t dt' E_1(t') \dot{C}(t-t') \quad (25)$$

This equation is now Laplace transformed into

$$\bar{\tau}_0 \tilde{C}(s) = \tilde{E}_1(s) \tilde{C}(s) \quad (26)$$

or

$$\tilde{C}(s) = \frac{\tau_0}{\ln(1+s) - \tau_0} \quad (27)$$

Here, use has been made of Eq. (9) and the identity

$\tilde{C}(s) = s\tilde{C}(s) - C(0) = s\tilde{C}(s) - 1$. $\tilde{C}(s)$ has a branch cut on the negative real axis from $s=-1$ to $s=-\infty$ and a pole at $s = \dot{\gamma}_0$, where

$$\dot{\gamma}_0 = e^{\tau_0} - 1 \quad (28)$$

is the steady state shear rate (Eq. (10)). The Laplace inversion of Eq. (27) is performed by deforming the integration contour to run from $-\infty$ slightly below the negative real axis, rounding the pole at $s = \dot{\gamma}_0$, and returning to $-\infty$ above the negative real axis. After standard manipulations one thus finds

$$\dot{C}(t) = \tau_0(1 + \dot{\gamma}_0)e^{\dot{\gamma}_0 t} + \tau_0 \int_1^{\infty} du e^{-ut} \frac{1}{[\ln(u-1) - \tau_0]^2 + \pi^2} \quad (29)$$

or finally, by integration with respect to time,

$$e^{J(t, \tau_0) \cdot \tau_0} = 1 + \tau_0 \frac{1 + \dot{\gamma}_0}{\dot{\gamma}_0} (e^{\dot{\gamma}_0 t} - 1) + \tau_0 \int_1^{\infty} du \frac{1 - e^{-ut}}{u} \frac{1}{[\ln(u-1) - \tau_0]^2 + \pi^2} \quad (30)$$

In the linear limit Eq. (30) reduces to

$$J(t, \tau_0 \rightarrow 0) = t + \int_1^{\infty} du \frac{1 - e^{-ut}}{u} \frac{1}{\ln^2(u-1) + \pi^2} \quad (31)$$

The creep compliance $J(t, \tau_0)$ of Eq. (30) is plotted in Fig. 4 in

a log-log plot for different values of $\dot{\gamma}_0$.

As a final example of the use of Eq. (7) consider the constrained recoil after a steady shear flow is interrupted at $t=0$ by suddenly removing the shear stress. We wish to calculate the so-called recoverable shear γ_∞ . Writing

$$\dot{\gamma}(t) = \begin{cases} \dot{\gamma}_0 & , t < 0 \\ -f(t) & , t > 0 \end{cases} \quad (32)$$

Eq. (7) implies for $t > 0$

$$0 = -\int_0^t dt' E_1(t') f(t-t') e^{-\int_{t-t'}^t f} + \int_t^\infty dt' E_1(t') \dot{\gamma}_0 e^{-\dot{\gamma}_0(t-t) - \int_0^t f} \quad (33)$$

or

$$\int_0^t dt' E_1(t') f(t-t') e^{\int_0^t f} = \dot{\gamma}_0 e^{\dot{\gamma}_0 t} \int_t^\infty dt' E_1(t') e^{-\dot{\gamma}_0 t'} \quad (34)$$

Defining $F(t) = \exp(\int_0^t f)$, Eq. (34) becomes

$$\int_0^t dt' E_1(t') \dot{F}(t-t') = \dot{\gamma}_0 e^{\dot{\gamma}_0 t} \int_t^\infty dt' E_1(t') e^{-\dot{\gamma}_0 t'} \quad (35)$$

The Laplace transform of Eq. (35) is

$$\tilde{E}_1(s) (s \tilde{F}(s) - 1) = \frac{\dot{\gamma}_0}{\dot{\gamma}_0 - s} (\tilde{E}_1(s) - \tilde{E}_1(\dot{\gamma}_0)) \quad (36)$$

or

$$\tilde{F}(s) = \frac{1}{s} \left[1 + \frac{\dot{\gamma}_0}{\dot{\gamma}_0 - s} \frac{\tilde{E}_1(s) - \tilde{E}_1(\dot{\gamma}_0)}{\tilde{E}_1(s)} \right] \quad (37)$$

The recoverable shear is determined from $e^{\gamma_{\infty}} = \lim_{t \rightarrow \infty} F(t)$. This limit is given by the residue at the pole at $s=0$ of Eq. (37), and one finds $\gamma_{\infty} = \ln(2 - \tilde{E}_1(\dot{\gamma}_0))$, or

$$\gamma_{\infty} = \ln(2 - \eta(\dot{\gamma}_0)). \quad (38)$$

In the two limits one has

$$\gamma_{\infty} = \begin{cases} \frac{1}{2} \dot{\gamma}_0 & , \dot{\gamma}_0 \ll 1 \\ \ln 2 & , \dot{\gamma}_0 \gg 1 \end{cases}. \quad (39)$$

$\gamma_{\infty}(\dot{\gamma}_0)$ is monotonously increasing which is also the case in experiment. Also like in experiment, γ_{∞} stabilizes on a recoverable shear of order one at high $\dot{\gamma}_0$.

3. DISCUSSION

The idea of this paper was to show that a simple constitutive relation with no adjustable parameters (except the overall scaling of time and viscosity) exists, which gives a qualitatively correct picture of shear viscoelasticity. Equation (7) was arrived at by requiring the Cox-Merz rule to be satisfied and that Eyring's nonlinear viscosity Eq. (8) is to be reproduced approximately. This ensures a nonlinear viscosity and a frequency-dependent linear viscosity which are both close to those observed in most experiments.

The choice of the linear relaxation modulus to be $E_1(t')$ may be justified instead from the box model, i. e. , the postulate of a uniform distribution of activation energies for microscopic motion. Consider the motion of a foreign microscopic particle in the liquid. Suppose the particle feels a spatially randomly varying potential energy, and that it moves by thermally activated hopping between the various potential energy minima. Then the linear mobility of the particle (the velocity divided by an external force acting on the particle), is given by

$$\mu(\omega) = \mu(0) \frac{i\omega}{\ln(1+i\omega)} \quad (40)$$

to a good approximation [6]. Assuming the Stokes law is valid for the particle, one has $\mu(\omega) \propto \eta^{-1}(\omega)$ which shows that the linear relaxation modulus of the liquid is $E_1(t')$ in this approximation.

Because the Cox-Merz rule is obeyed by the model it is not surprising that the Gleissle mirror relation [1] is also satisfied: The linear limit of η^+ from Eq. (16) is

$$\lim_{\dot{\gamma}_0 \rightarrow 0} \eta^+(t, \dot{\gamma}_0) = t E_1(t) - e^{-t} + 1 \quad (41)$$

Gleissle's mirror relation states that $\eta(\dot{\gamma})$ is equal to this limit evaluated at $t = 1/\dot{\gamma}$, thus

$$\eta(\dot{\gamma}) = \begin{cases} 1, & \dot{\gamma} \ll 1 \\ (1 - C + \ln \dot{\gamma}) / \dot{\gamma}, & \dot{\gamma} \gg 1 \end{cases} \quad (42)$$

where $C=0.577\dots$ is Euler's constant. A comparison of Eqs. (10) and (42) shows that the mirror relation is indeed satisfied to a good approximation.

The constitutive relation Eq. (7) reproduces most qualitative features of shear viscoelasticity. (An exception is the overshoot usually observed in η^+ as function of time, where the present model predicts η^+ to increase monotonously to the steady state value.) The fact that qualitative features of experiment are generally reproduced is not surprising, given the similarity between the present model and the Wagner model which is well-known to give a satisfactory description of experiment. The main difference between the present model and Wagner's model is that the damping function is here $\exp(-\int_{t-t'}^t |\dot{\gamma}|)$ whereas Wagner has $\exp(-\int_{t-t'}^t \dot{\gamma})$. The appearance of $\int_{t-t'}^t |\dot{\gamma}|$ in the damping function is a simple way to incorporate irreversibility [7] into the model, i. e., to reflect that segments are lost irreversibly

during any deformation. A possible objection to this kind of damping term is that, for a periodic shear $\gamma = \gamma_0 \sin(\omega t)$, one might expect nonlinearity to set in at high frequencies even at very small amplitudes (because the damping apparently is a function of $\gamma_0 \omega$, and not of γ_0). This, however, is not correct: Suppose the worst possible case of the nonlinearity, i. e., put the damping function equal to $\exp(-\omega \gamma_0 t')$ in Eq. (7). Then the response is

$$\begin{aligned} \bar{z}(t) &= \gamma_0 \omega \int_0^{\infty} dt' E_1(t') \cos(\omega(t-t')) e^{-\omega \gamma_0 t'} \\ &= \gamma_0 \omega \left[\cos(\omega t) \operatorname{Re} g - \sin(\omega t) \operatorname{Im} g \right] \end{aligned} \quad (43)$$

where

$$g = \int_0^{\infty} dt' E_1(t') e^{-(i\omega + \gamma_0 \omega)t'} = \frac{\ln(1+x)}{x}, \quad x = i\omega + \gamma_0 \omega \quad (44)$$

At a fixed ω the onset of nonlinearity may be estimated from

$$\gamma_0 \omega \approx \left| \frac{g(x)}{g'(x)} \right|_{x=i\omega} \quad (45)$$

which is the criterion for the first order term being equal to the zero'th order term in the Taylor expansion of g as function of γ_0 . Equation (45) leads to

$$\gamma_0 \omega \approx \omega \left| \frac{\ln(1+i\omega)}{\frac{i\omega}{1+i\omega} - \ln(1+i\omega)} \right| \quad (46)$$

It is now easy to see that whenever $\omega \gg 1$ the onset of

nonlinearity takes place for γ_0 of order one. For $\omega \ll 1$, on the other hand, the onset of nonlinearity is at $\gamma_0 \approx \omega^{-1}$ corresponding to a maximum shear rate of order one in the periodic variation.

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FIGURE CAPTIONS

Fig. 1: Log-log plot of various quantities characterizing the model. In this figure, as everywhere in the paper, dimensionless time, stress, and viscosity are used, the latter quantity normalized so that $\eta_o(\omega=0)=1$. The figure shows: (1) The predicted nonlinear viscosity as function of $\chi=\dot{\gamma}$ (•) (Eq. (10)), (2) Eyring's nonlinear viscosity as function of $\chi=\dot{\gamma}$ (Δ) (Eq. (8)), (3) $|\eta_o(\omega=\chi)|$ (∇) (Eq. (11)), and (4) the real (+) and the imaginary (\times) part of $\eta_o(\omega=\chi)$. A comparison of the • and Δ points shows that Eyring's viscosity, which is known to give a good fit to many experiments, is reproduced reasonably well by the model. Comparing the • and the ∇ points shows that the Cox-Merz rule is obeyed, though not quite accurately in the transition region. The real and imaginary parts of $\eta_o(\omega)$ looks much like in experiment.

Fig. 2: Stress growth upon inception of a steady shear flow with shear rate $\dot{\gamma}_o$. The quantity $\eta^+(t, \dot{\gamma}_o)$ given by Eq. (16) is plotted as function of time for: (a) $\dot{\gamma}_o=0.1$ (reflecting the linear limit), (b) $\dot{\gamma}_o=3$, (c) $\dot{\gamma}_o=10$, and (d) $\dot{\gamma}_o=30$. Like in experiment, $\eta^+(t, \dot{\gamma}_o)$ follows the linear $\eta^+(t)$ for short times while it stabilizes for large t at the nonlinear viscosity, a stabilization which takes

place sooner the larger $\dot{\gamma}_0$ is. The overshoot of $\eta^+(t, \dot{\gamma}_0)$ often seen in experiment is not reproduced by the model.

Fig. 3: Stress growth after cessation of a steady shear flow with shear rate $\dot{\gamma}_0$. The figure shows the quantity $\eta^-(t, \dot{\gamma}_0)/\eta(\dot{\gamma}_0)$ as function of time, where η^- is given by Eq. (20), for: (a) $\dot{\gamma}_0 = 0.1$ (reflecting the linear limit), (b) $\dot{\gamma}_0 = 3$, and (c) $\dot{\gamma}_0 = 30$. As in experiment, $\eta^-(t, \dot{\gamma}_0)$ decreases to zero as $t \rightarrow \infty$ faster the larger $\dot{\gamma}_0$ is.

Fig. 4: Creep compliance $J = \gamma(t)/\bar{\tau}_0$ where J is given by Eq. (30), plotted as function of time for: (a) $\dot{\gamma}_0 = 0.1$ (reflecting the linear limit), (b) $\dot{\gamma}_0 = 1$, (c) $\dot{\gamma}_0 = 10$, and (d) $\dot{\gamma}_0 = 100$, where $\dot{\gamma}_0$ is related to $\bar{\tau}_0$ by Eq. (28).

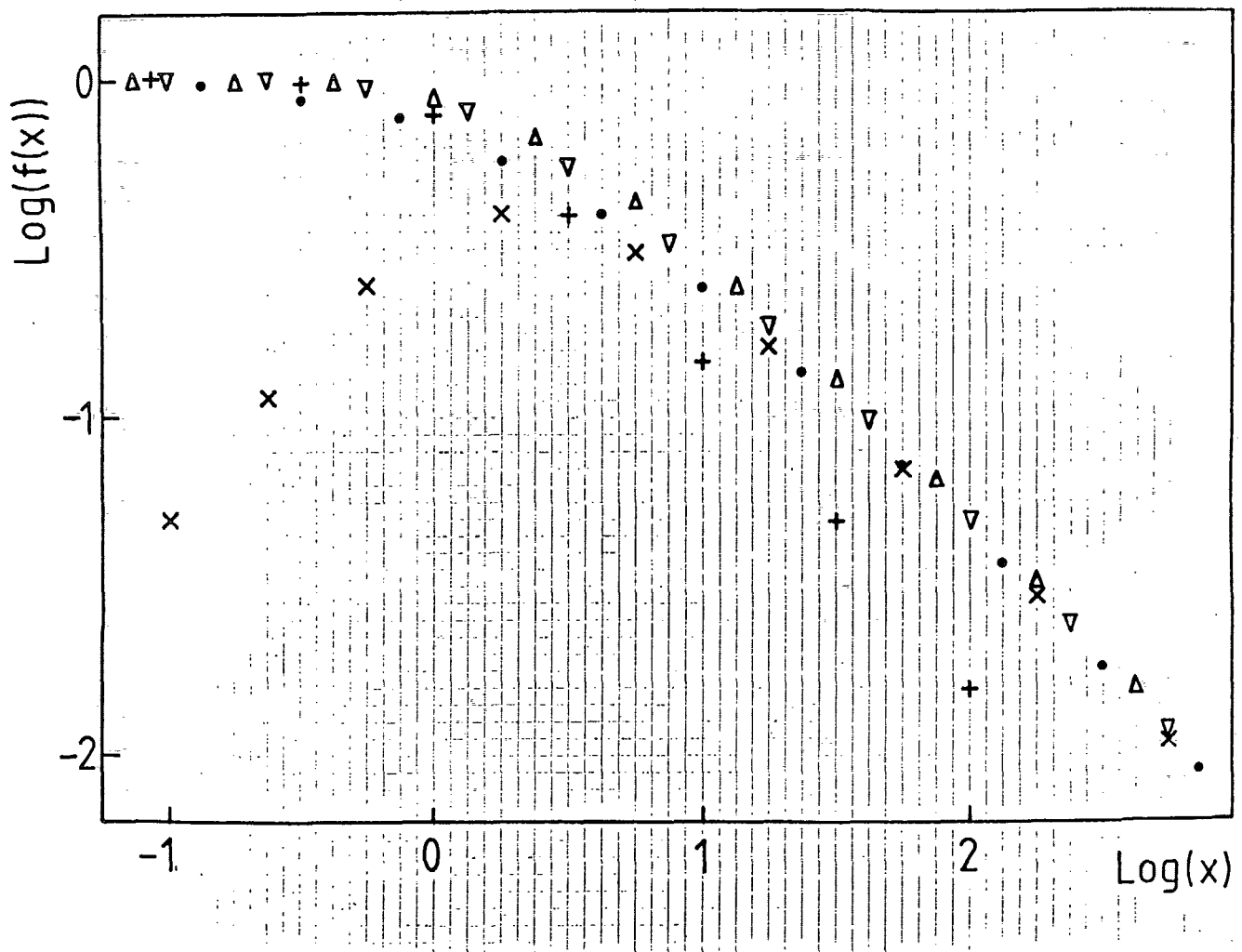


Fig. 1

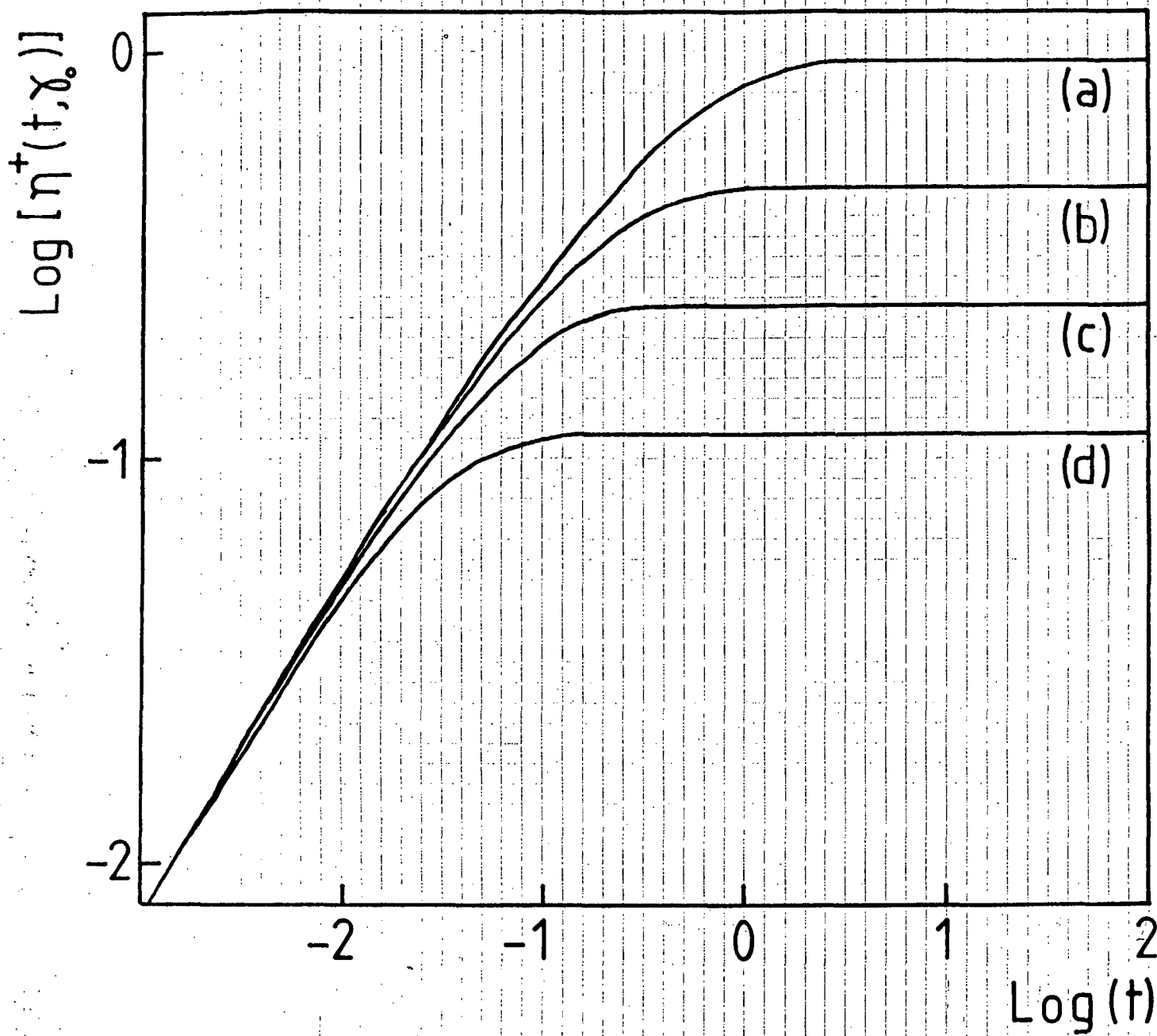


Fig. 2

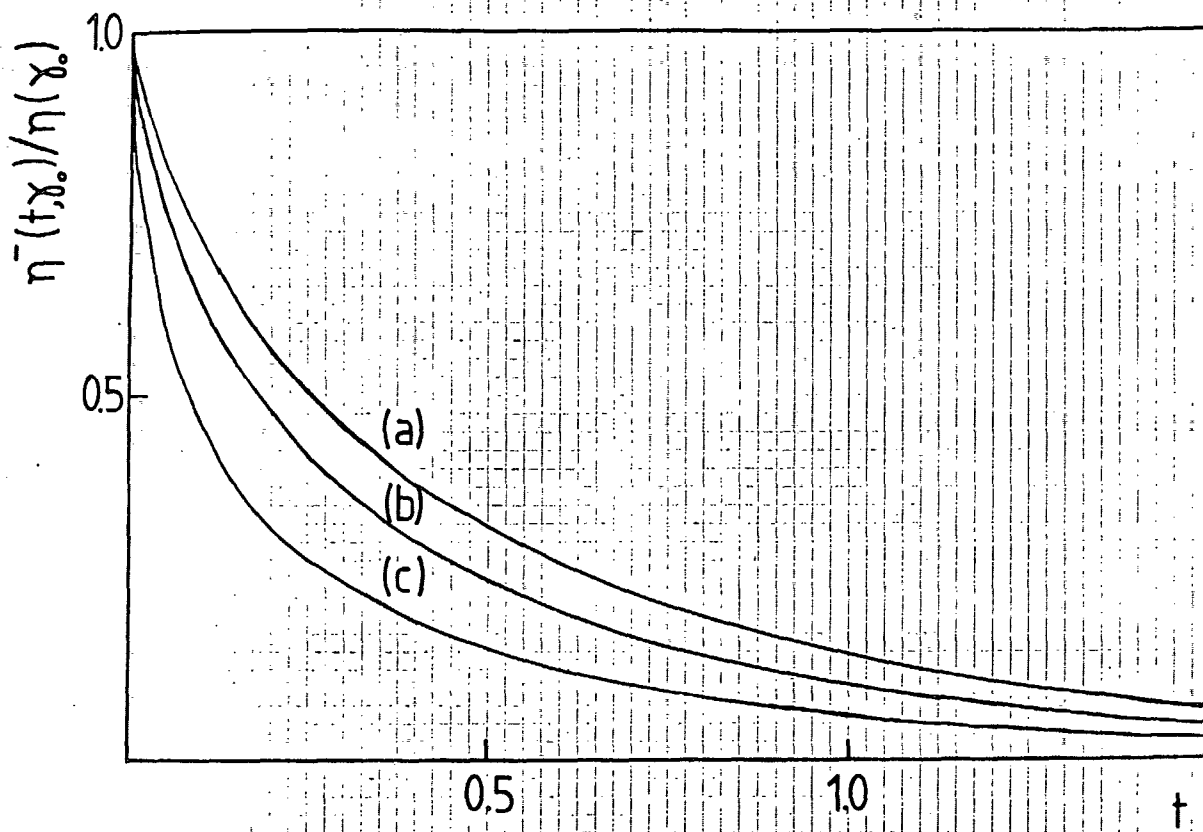


Fig. 3

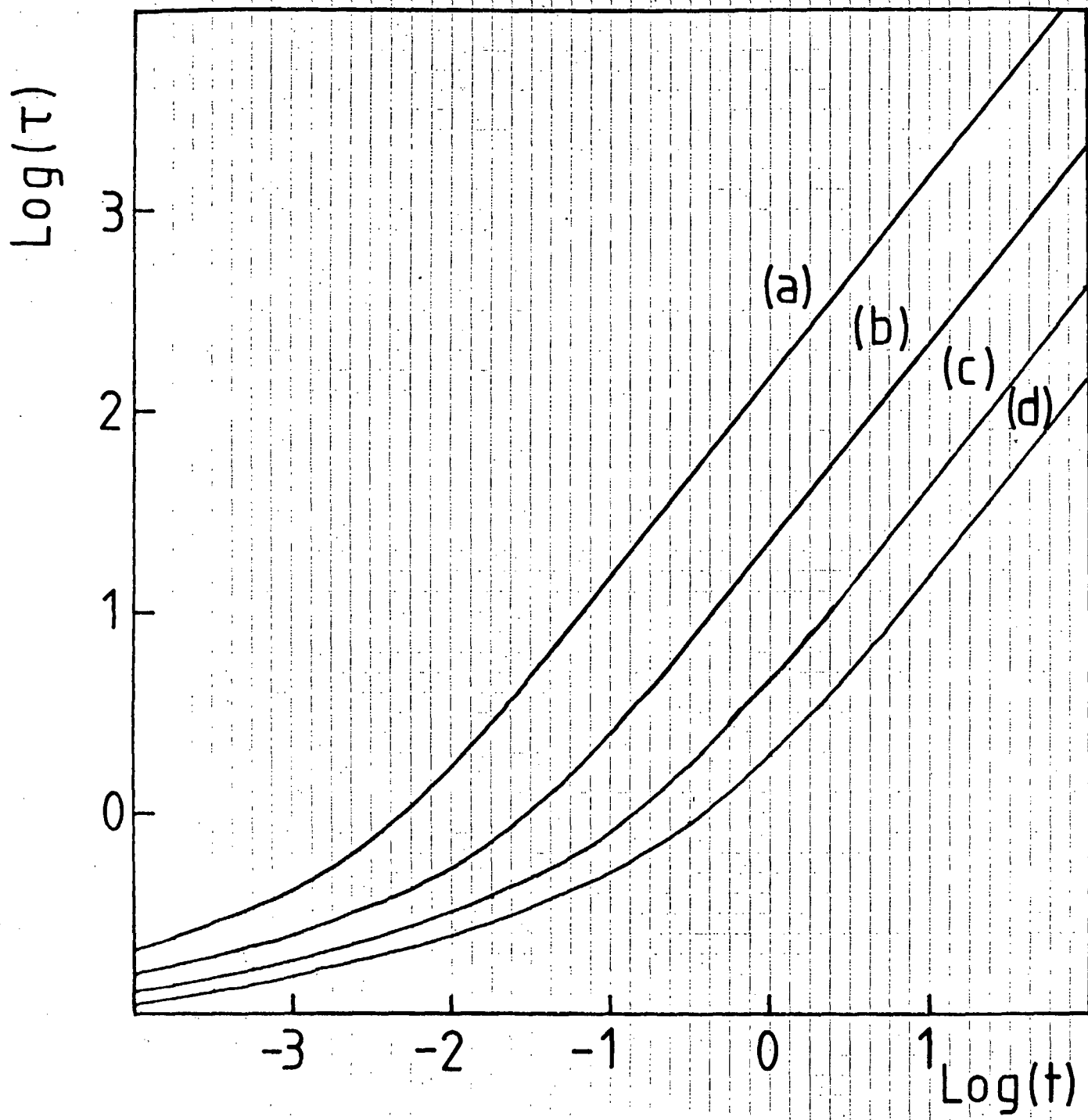


Fig. 4

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