

## Rheology of uncured magnetorheological elastomers

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**Abstract.** The aim of this paper is to study rheological properties of uncured magnetorheological elastomers comprising iron particles dispersed in silicon elastomer in relation to particle rearrangement by external magnetic field into oriented structures as this process is strongly affected by viscosity. Studied systems vary in used filler volume concentration (0 to 30 vol. %). From measured flow curves flow consistency index is extracted first by fitting experimental data to Herschel-Bulkley model followed by applying two concentration dependency models (Maron-Pierce and Krieger-Dougherty) to normed consistency. Results and model predictions are discussed.

### 1. Introduction

Magnetorheological composites with sufficiently low viscosity of an uncured matrix/binder allow for filler preferential rearrangement therein thus creating unique inner structure material of physically interesting properties [1, 2]. Provided the filler, to be arranged, is ferromagnetic, one can employ external magnetic field to do so. Depending on filler loading and inherent viscosity of the mixture filler particles are drawn into clusters along the direction of applied magnetic field. These clusters tend to be chain-like for dilute systems but gradually composite loses this morphology with increasing filler concentration as steric factors and substantially increased viscosity prevent large distance filler translation [3].

Thus created structured magnetic elastomers can be employed for instance in piezosensors as the conductivity of chain-like aggregates is sensitive to any deformation stemming from external stimuli in form of applied pressure [3, 4]. However composites with organised inner structure can be beneficial even in other fields such as shielding of electromagnetic radiation. Magnetic particles filled elastomers have been utilised in shielding applications for years due to flexibility and decent shielding properties. Originally uniform spatial distribution of filler particles allows for changes only via concentration which still undoubtedly remains the main factor affecting shielding efficiency. Nevertheless filler structuring inside the elastomer can considerably alter how such material interacts with electromagnetic wave be it reflection, absorption or transmission [5-7].

Arrangement into chain-like clusters by the effect of magnetic field is crucially affected by the loading and becomes increasingly difficult for highly concentrated system. This is partly due to spatial reasons but also owing to increase in viscosity. Viscosity of suspensions and its dependence on filler depends on many factors such as particle shape, particle size distribution but mainly on filler volume loading. The dependence of suspension viscosity versus its volume loading is theoretically described with several models depending on concentration region. Dilute systems up to approximately



2 vol. % exhibit linear dependence between filler volume loading and corresponding viscosity of the mixture and are well approximated by Einstein equation:

$$\eta = 1 + B\phi \quad (1)$$

where  $B$  is intrinsic viscosity (= 2.5) and  $\phi$  is volume fraction of the filler.

In the semi-dilute region spanning from 2 to about 25 vol. % dynamic viscosity shows higher order dependence on filler concentration but still remains rather Newtonian.

$$\eta = 1 + B\phi + B_1\phi^2 + \dots \quad (2)$$

where  $B = 2.5$  and  $B_1 \in \langle 7.35; 14.1 \rangle$ .

Finally, suspensions with loading above 25 vol. % have strongly non-Newtonian behaviour with rapid growth of viscosity on filler concentration. In this concentrated regime two models are often used to approximate experimental data, namely Maron-Pierce:

$$\eta = \left(1 - \frac{\phi}{\phi_m}\right)^{-2} \quad (3)$$

and its more flexible modification done by Krieger-Dougherty:

$$\eta = \left(1 - \frac{\phi}{\phi_m}\right)^{-B\phi_m} \quad (4)$$

where  $\phi_m$  is maximal filler fraction and  $B$  is a fit parameter, i.e. Krieger-Dougherty model has two parameters compared with Maron-Pierce [8].

In this study we have tried to capture the effect of magnetic filler loading on viscosity of uncured magnetorheological systems, which in turn strongly affects the tendency of the filler particles to be rearranged by magnetic field into structures oriented parallel to it.

## 2. Experimental

### 2.1. Materials & Suspensions preparation

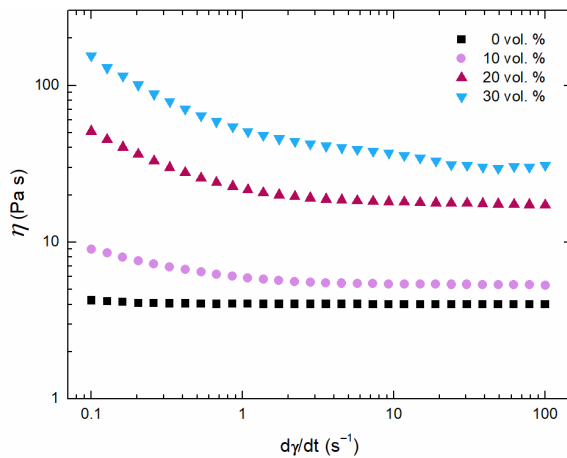
Prepared uncured magnetorheological elastomer suspensions (i.e. without addition of a curing agent) comprised silicon elastomer (Sylgard 184, DowCorning, USA) matrix filled with iron particles (carbonyl iron, CN grade, BASF, Germany). Calculated amount of filler corresponding to the chosen concentration (10, 15, 20, 25 and 30 vol. %) was transferred into silicon elastomer and mixed (450 rpm) for 5 min by a vacuum mixer in order to prevent introduction of air into the mixture.

### 2.2. Rheology

Viscosity of the suspensions was performed under controlled shear rate mode with a rotational rheometer Physica MCR 502 (Anton Paar GmbH, Austria) with a Peltier temperature unit using parallel-plate measuring system with a diameter of 25 mm (PP25) and a gap of 0.5 mm. All measurements were performed at 25 °C.

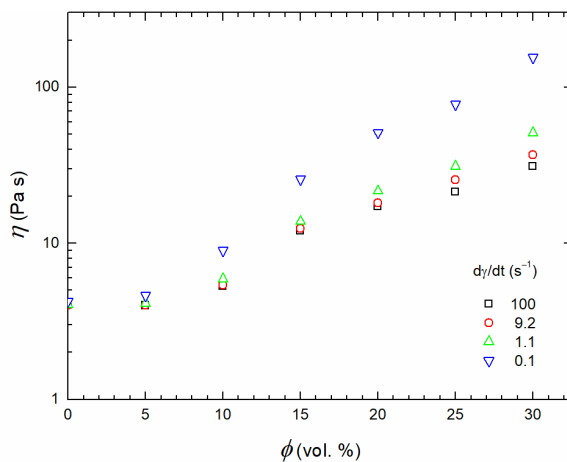
## 3. Results and discussion

To provide a complete characterisation also the viscosity of carrier medium, i.e. silicon elastomer, was measured. Silicon's dynamic viscosity expectedly observed Newtonian behaviour with constant value of 4 Pa s in the whole of investigated shear rate region (Fig. 1). However all the other systems including the 5 vol. % (excluded from the Fig. 1 for better clarity) showed pseudoplastic behaviour with increasing shear rate which at the end of measured rates levelled off into a constant value. Higher loadings saw this trend to be more pronounced. Due to this relaxation of viscosity from theoretical first Newtonian plateau into the second one it does matter for which shear rate one evaluates the viscosity increase with filler concentration. Nevertheless in our case related to investigation of filler particles rearrangement under the effect of magnetic force one can assume to deal with rather low shear rates probably not exceeding several  $s^{-1}$ . Still we plotted the rise of suspensions' viscosity with filler incorporated for several shear rates covering the whole shear rate range measured (Fig. 2).



**Figure 1.** Flow curves of variously concentrated uncured magnetorheological elastomers

It can be seen that viscosity relaxation indeed influences the concentration dependence as the shear rate of  $0.1 \text{ s}^{-1}$  clearly exhibits character different from the other three shear rates. But even so one cannot but notice sharp break in viscosity increase with suspension volume concentration at 5 vol. % in agreement with theoretical predictions as this concentration point lies on the boundary of dilute/semi-dilute systems.



**Figure 2.** Dependence of dynamic viscosity of uncured magnetorheological elastomers on filler volume concentration

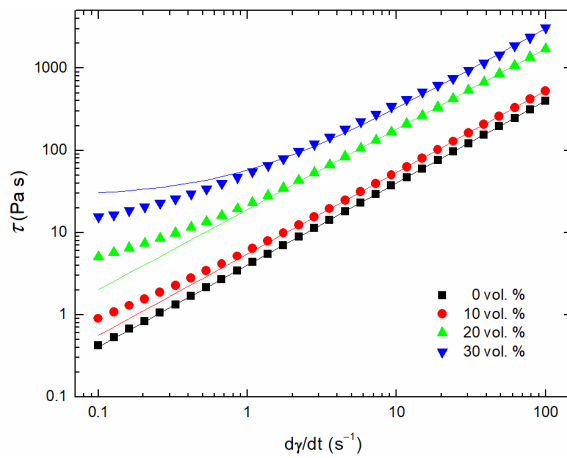
In order to overcome the effect of shear rate we decided to fit the experimental dependence of shear stress ( $\tau$ ) versus shear rate ( $\dot{\gamma}$ ) by Herschel-Bulkley model [9]:

$$\tau = \tau_0 + K\dot{\gamma}^n \quad (5)$$

where  $\tau_0$  (Pa) is yield stress,  $K$  ( $\text{Pa s}^n$ ) flow consistency index and  $n$  flow behaviour index.

Even though model partially fails to properly approximate the data below  $1 \text{ s}^{-1}$  it gives very satisfactory results in high shears (Fig. 3). Obtained values of parameters in Herschel-Bulkley model (Tab. 1) namely flow consistency index ( $K$ ) was subsequently normed to carrier liquid (silicon elastomer) extracted consistency ( $K_0 = 4.04$ ):

$$K_{\text{norm}} = \frac{K}{K_0} \quad (6)$$

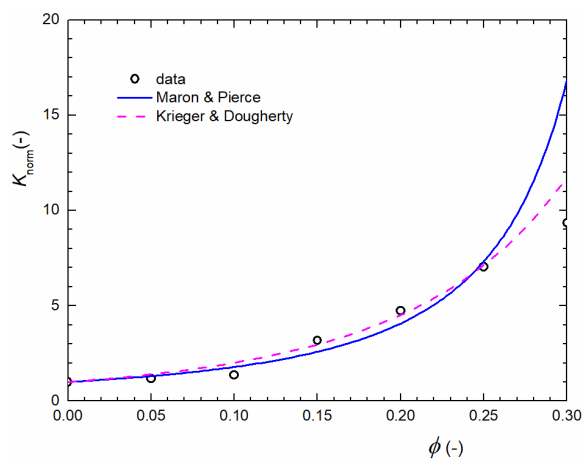


**Figure 3.** Flow curves of variously concentrated suspensions approximated by Herschel-Bulkley model

**Table 1.** Fitting parameters of Herschel-Bulkley model for measured flow curves

$\Phi$	$K$	$n$
0	4.04	0.998
0.05	4.71	0.995
0.1	5.48	0.993
0.15	12.80	0.986
0.2	19.15	0.977
0.25	28.44	0.955
0.3	37.75	0.949

Finally normed consistency ( $K_{\text{norm}}$ ) for each concentration point, with the exception of  $\Phi = 0.3$ , which was identified as an outlier, was fitted with Maron-Pierce (eq. 3) and Krieger-Dougherty (eq. 4) models [8]. Even though both models fit the experimental data reasonably well (Fig. 4), Krieger-Dougherty equation due to higher flexibility in the form of an extra fitting parameter capture the character of the data more precisely.



**Figure 4.** Normed consistency flow index of suspensions as a function of their filler fraction fitted by models

Both models failed to provide sensible results when concentration point  $\Phi = 0.3$  was included into calculation, especially parameter  $\Phi_m$  in Krieger-Dougherty model tended to diverge to given upper limit of parameter constraint, thus it was not taken into account, however for the sake of completeness it was left in the plot. Despite the fact that both used models feature the same parameter  $\Phi_m$  each

model yielded different value; thus least square method for Maron-Pierce model gives 0.37 while Krieger-Dougherty model worked best with substantially higher value of 0.79. This is nonetheless caused by the presence of additional parameter  $B$  (best fit for  $B = 6.54$ ) which together with  $\Phi_m$  modify in Krieger-Dougherty equation otherwise fixed exponent of  $-2$  in Maron-Pierce equation.

In order to further verify predictions of both models for real systems more concentrated suspensions should be investigated as well as other effects such as tendency of filler to aggregation or its particle size distribution.

#### 4. Conclusions

Preparation of structured magnetorheological elastomers by the application of external magnetic field during curing process to achieve magnetic particles arranged into chain-like clusters oriented along the direction of magnetic field have several challenges one of which is the gradually increasing viscosity of the system with filler loading. To this end we prepared a set of variously concentrated (0 to 30 vol. %) uncured magnetorheological systems comprising components typically used in magnetorheology and measured their flow characteristics. Obtained flow curves were fitted by Herschel-Bulkley model to extract consistency flow indexes whose norms were approximated by two models, Maron-Pierce and Krieger-Dougherty, predicting the rise of suspension's viscosity with filler fraction. Both models provide satisfactory results in the measured concentration range with Krieger-Dougherty following the experimental data more closely compared to slightly looser character of Maron-Pierce fit. Their different character can be seen outside the fitted concentration range where they tend to increase with filler loading at considerably different rates. Viscosity measurement shows strongly convex (non-linear) dependence on filler concentration confirming only very difficult filler rearrangement at higher loadings namely 30 vol. %.

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