Silicon (2019) 11:1287–1293
https://doi.org/10.1007/s12633-018-9921-1

ORIGINAL PAPER

Analysis of the Dynamic Behavior of Magnetorheological Elastomer Composite: Elaboration and Identification of Rheological Properties

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Received: 26 September 2017 / Accepted: 25 May 2018 / Published online: 20 June 2018

Abstract

The present work is devoted to experimental analysis of the magnetorheological elastomer composite behavior under dynamic loading. The elastomer is charged to 40% of ferromagnetic particles. The characterization of the rheological properties was performed and the relation between the loads and the applied magnetic field has been studied. The results found show that this composite presents strong energy dissipation, further accentuated by the structure and the magnetic field.

Keywords Magnetorheological elastomer · Silicon · Dynamic loading · Rheological properties · Energy dissipation

1 Introduction

The elastomer exhibits a viscoelastic behavior when dynamic loading is applied; it presents at the same time the properties of elastic solid, and those of viscous fluid [1, 2]. The viscoelastic properties of a composite structure can be controlled by means of a magnetic field [3, 4]. The aim is to evaluate the significant energy dissipation of the composite structure, further increased in the presence of magnetic field [5]. The 1980s saw the birth of an interest for the materials with various properties under the influence of an external factor (temperature, electric or magnetic fields) including the magnetorheological elastomer (MRE) [6]. These materials (MRE) are composed of magnetic particles polarizable which are dispersed in a matrix made generally in silicone oil. Today, several scientific laboratories conduct research on the MRE. Usually elastomers for flexible silicone or polyurethane are used for polymer matrix. They are filled with a significant part of magnetic particles, often 30% of volume. To reduce the amount of magnetic particles, a vulcanization process forced by a magnetic field has been used in order to increase their effectiveness. The MRE made of flexible matrices of silicone or polyurethane show a significant response to magnetic fields but their low mechanical properties prevent them to be used in engineering [7]. It is expected that these materials, although they are in the development phase, will be very useful for solving some problems of vibrations, which are current problems for the construction and the use of machines or systems. The magnetorheological effect and the damping properties are basic for the applications of MRE. They are used in particular to achieve the dampers, the bushings of suspensions and magnetostrictive materials [8]. To determine magnetic parameters, it is necessary to design a test bench as well as to prepare the test allowing getting desired characteristics. In order to obtain the hysteresis curve of the MRE, the following experiments were performed [9, 10]. In these last years, the scientific community concentrate on the knowledge of the rheological behavior. Valery P et al. [11] have studied the isolation from vibrations by the use of magnetorheological elastomer. This experimental work mainly presents the parameters of the active shock absorber which is more important. Mirosław Bocian et al. [12] worked out a mechanical structure based on a magnetorheological elastomer; this elastomer has been designed for the dissipation of energy and the mitigation of the vibratory movements from an excitation of impact. An adequate mathematical model was derived by Mateusz Kukla et al. [13] this work presents the results of the behavior analysis in compression and the effect of the static magnetic field It also presents an attempt to use of rheological models to describe the

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MRE. The influence of the γ radiation on the shear modulus of the magnetorheological elastomer was studied [14]. The experimental results show that the initial shear modulus and the shear modulus induced by magnetic field increase in the first, and then decrease with the increase of the radiation dose. Two factors are considered to explain the experimental results. One is the reaction of crosslinking and degradation induced by radiation, the other is the change of the magnetization of the saturation of the particle of iron carbonyl. Schümann et al. [15] carried out a mechanical characterization at several time scales, in order to obtain an overview on the electrical and mechanical behavior at short and long term of this new material. The results found show a complex resistivity behavior over several time scales, which is sensitive to the magnetic fields and to the strain rate. Agirre-Olabide et al. [16] created a new magnetodynamic compression technique to measure the magneto-viscoelastic properties of magnetorheological elastomers (MRE) at high-frequencies. Isotropic MRE filled with carbonyl iron powder were synthesized, and three volumetric particle proportions were studied: 0%, 15% and 30%. The viscoelastic properties were calculated by the use of stress-strain diagrams for each condition of frequency and strain amplitude. Bunoiu [17] manufactured hybrid magnetorheological elastomers (HMREs) from sponges filled with magnetorheological suspensions (MRS) constituted of silicone oil (SO) and carbonyl iron in volume fractions (Φ) of 10%, 30% and 60%. They showed that the dielectric and elastic behavior of the HMRE is influenced by the applied magnetic field and also the volume fraction Φ. Schümann [18] in his work used X-ray microtopography to analyze the microstructure of particles. In addition, a characterization of the microstructure of the particles in the presence of magnetic fields has been made. A significant impact of magnetic field and deformation on particle rotation and radial distribution has been clearly observed. Zhenlong [19] used the finite element method to analyze structures with a magnetorheological elastomer core. The results found show that the MRE core can be controlled by the application of an external magnetic field; in addition, they can be adjusted by changing the thickness of the latter. Marc-André et al. [20] in their work, they analyzed the effective response of several samples of MRE with respect to the individual impact of microscopic morphology and macroscopic shape. This latter approach allows multi-scale computational characterization of magnetorheological elastomers with arbitrary shapes and microstructures. As examples of prototypes, they analyzed two-dimensional specimens with rectangular and elliptical shapes. In addition, they presented a generic analytical approach for shape effects, which is based on magneto-mechanical tensions acting on the surface of MRE specimens. The results show that the analytic approach is able to predict fundamental stress states and strain tendencies, which have also been observed in numerical simulations.

In this work we have studied the influence of the variable applied magnetic field on the rheological properties of an elastomer charged at 40% of iron particles. Firstly, the development of the sample of magnetorheological elastomer is presented. Secondly, the influence of the magnetic field on the adjustable dynamic properties of the MRE specimen is studied experimentally using dynamic viscoanalyser and the obtained results for different magnetic field intensities are presented.

2 Theoretical Modeling and Experimental Analysis

2.1 Theoretical Modeling

In this work, we will see that the generalized Maxwell model is suitable for describing the mechanical behavior of our elastomer; this model consists of a spring and N models of Maxwell arm assembled in parallel. The elasticity moduli are denoted by $G_0, G_1, G_2, \ldots, G_n$, while viscosity coefficients are designated by $\eta_1, \eta_2, \ldots, \eta_n$.

Considering two functionally graded layers bonded by viscoelastic elastomer wich can be modeled by Maxwell-Wiechert model [8]. This model contains a series of spring-dashpot units and a hookean spring. The time-dependent shear modulus $G(t)$ of the viscoelastic elastomer varies with time and can be expressed as Prony series

$$G(t) = G_\infty + \sum_{i=1}^{N} G_i e^{-t/\tau_i} \tag{1}$$

Where G_∞, G_i and τ_i are the long-term shear modulus, the relaxation shear moduli and the relaxation time.

The strain of the model is the sum of the strains of the two elements, represented by the spring element and the dashpot such as:

$$\tau = E\varepsilon + \gamma\dot{\varepsilon} \tag{2}$$

Based on Boltzmann superposition principle, the shear stress $\tau(x, t)$ can be expressed as:

$$\begin{aligned}\tau(x,t) &= G(t)\gamma(x,0) + \int_0^t G(t-\xi)\frac{\partial\gamma(x,\xi)}{\partial\xi}d\xi \\ &= G(t)^* d\gamma(x,t) = \gamma(x,t)^* dG(t)\end{aligned} \tag{3}$$

Where the $(*)$ denotes convolution. The equation above describes relaxation constitutive relationship. After Fourier transform, this can be expressed as

$$\tau(x,\omega) = i\omega G(\omega).\gamma(x,\omega) \tag{4}$$

Where $G(\omega)$ is the Fourier transform of shear modulus $G(t)$ and i denotes $\sqrt{-1}$, and this equation can also be written as

$$\tau(x,\omega) = G^*(\omega).\gamma(x,\omega) \tag{5}$$

To take account of the duality between viscosity and elasticity, we frequently use complex numbers (two components) when a material is subjected to a dynamic solicitation, the complex modulus $G^*(t)$ for a shear solicitation, is given by

$$G^* = G' + iG'' = G'(1 + i\eta) \tag{6}$$

G' is the real part, called storage modulus, that characterizes the rigidity of the elastomer and G'' the imaginary part, called loss modulus, which characterizes the viscous behavior.

The loss factor or damping factor is written as

$$\tan(\delta) = \frac{G''}{G'} = \eta \tag{7}$$

2.2 Experimental Analysis

The objective of this work is devoted to understanding the role of microstructure on the macroscopic response of the elastomer composite when it is subjected to magneto-mechanical loading conditions. The first part of the work aims to set up the elastomer composite manufacturing process from different ingredients (silicone oil, magnetizable iron particles, RTV141); the second part consists of determining the rheological characteristics as well as the strength of attraction between the micron-size ferromagnetic particles as a function of the excitation frequencies.

2.2.1 Choice of Constituents

a. Matrix The realization of a structured composite material can not be done in any conditions. First, the elastomer must have good mechanical properties, but also a low viscosity before crosslinking to facilitate the dispersion and structuring of the charges. Crosslinking at room temperature is a clear advantage because it facilitates the implementation of structuring in the field.

The choice fell on a silicone elastomer, marketed by Rhodorsil RTV 141, this polymer has a sufficiently low viscosity so that the dispersion of 30% of filler remains easy, but at the expense of its mechanical properties: very friable, with long degassing, rather, we will use its transparency and reserve for these composites rather qualitative analyzes. These characteristics are given in Table 1.

Table 1 Characteristics silicone elastomer RTV 141

	Primary	Catalyst	Mixed
Viscosity (Pa.s)	3.5	0.65	4
Young's modulus (kPa)			700
Elongation at rupture			120%
Release time (h)			4(60°) or 2(100°)
Density			1.02
Color	Transparent	Transparent	Transparent

b. Charges Particles that are too small make it difficult to create a good structure. To facilitate characterization, their morphology will have to be spherical and not very polydisperse. The choice of particles was fixed on Prolabo Normapur iron with high purity (99.5%), Table 2.

2.2.2 MRE Material and Implementation

A Dynamic Mechanical Thermal Analyzer (DMTA) machine is used to perform dynamic tests on small samples by varying the frequency. In our case we have determined the rheological properties of MRE using dynamic mechanical analysis (MetraviB DMA +450). The Metravib DMA +450 consists of a very rigid frame and a movable punch. The punch is connected to two coils (carrying an alternating current) in a magnetic field. The current variations in the coil result the vertical displacements of the punch due to the electromagnetic force induced by the current; this allows performing the tests to the excitation frequency of 1000Hz. In order to register the forces and displacements, there are 4 sensors in the device. A magnetic displacement sensor that operates between 1Hz to 100Hz frequencies and an accelerometer operating until a frequency of 1000Hz. By integrating the signal of the accelerometer, it can plot the displacement of the punch.

2.2.3 Elaboration Process of the Elastomer

The elastomer is elaborated by the following steps

1- Mixing the silicone oil and the RTV141A polymer in a glass bowl and proceed with a manual mixing during 15 minutes to obtain an elastomeric gel with good homogenization. A second bowl is used, containing a quantity of iron particles of micrometric size, with average diameter of 1.8-2.3 μm, to loading the elastomer.

2- A quantity of this gel obtained by silicone and RTV141A is mixed during 30 minutes with a quantity

Table 2 Characteristics of iron 99.5

Fe%	Insoluble impurities
99.5	0.1

of iron particles until obtaining a homogeneous paw. By this process, an elastomer charged with 40% of ferromagnetic particles is elabored.

3- In order to have a healthy structure for the experimentation, the degassing of the obtained paw under vacuum during 10 minutes to eliminate air bubbles infiltrated during the mixing is performed. Because of the temperature influence, which can cause the reticulation of the elaborate paw, the obtained elastomer is hermetically preserved at low temperature (0 °C).

After the preparation of the experimental device (switching on the Metravib DMA + 450, the rectangular inner form aluminum mold and the two coils generating the magnetic field), the RTV 141B, which acts as a catalyst, is added to the paw, for increase the constituents adhesion in this latter; then injected in the aluminum mold so that it fills its entire volume. This must be done rapidly to avoid the reticulation. 10 specimens are elabored and each sample has a rectangular shape of 35 mm of length, 25 mm of width, and 2 mm of thickness.

Experimental tests depicting the shear strain on an elastomer sample are conducted at variable frequency from 0 to 100 Hz with and without the magnetic field. MER material is maintained under these experimental conditions during 24 hours and at the ambient temperature of 27 °C until the reticulation.

Without a magnetic field, ferrous particles are randomly distributed in nonmagnetic silicone to form a magnetorheological elastomer (top), Fig. 1a. Once a magnetic field is applied, the particles align with the magnetic field to form chains (Fig. 1b), the viscosity increase, dramatically, in the perpendicular direction to the field direction (bottom).

Table 3 Constituents of the magnetorheological elastomer

Time of reticulationen	$m_{\text{SiliconOil}}$	$m_{\text{RTV(A)}}$	m_{Fe}	$m_{\text{RTV(B)}}$
Charged elastomer to 40% iron particles				
28h	1.456g	1.253g	8.900g	0.254g

The ingredients of specimen in magnetorheological elastomer charged to 40% of iron particles of its total volume are given in Table 3.

3 Results and Discussion

Experimental results obtained by dynamic analysis (Dynamic Viscoanalyseur DMA+450) of MRE charged at 40% of the iron particles exposed under different intensities of the magnetic field are given by Table 4. These results show that the values found are sufficiently accurate with a small error.

The curves modulus-strain of isotropic composites charged to 40% with and without magnetic field is compared (Fig. 2a and b). On this figure, it is observed that the storage modulus and the loss modulus decrease as a function of the increase of shear strain, we distinguish, a sudden change of the storage modulus and the loss modulus for a shear deformation less than 4%, then a slow change for a shear strain greater than 4%.

The explanation of this stiffening of the material submitted to a field is the following: at the microscopic level, the magnetic field creates an attractive inter-particular

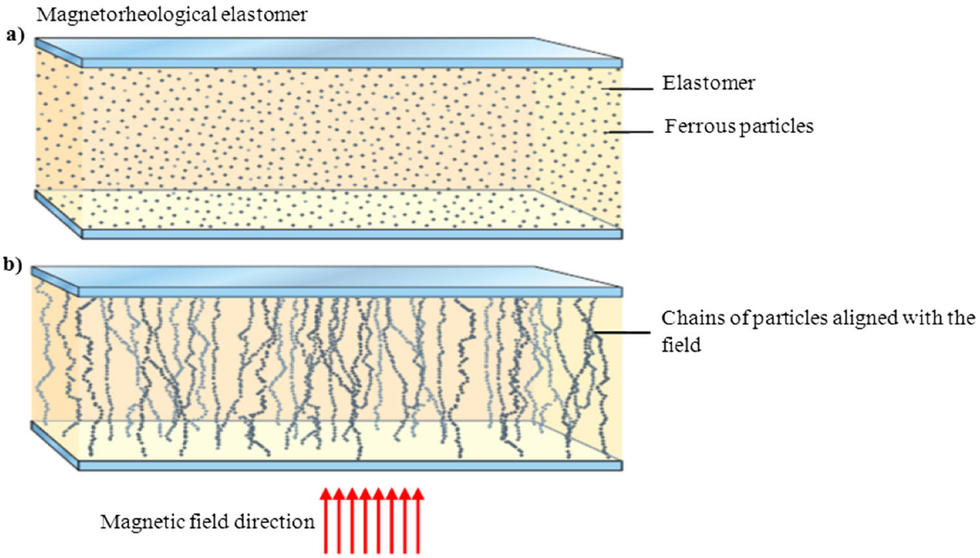

Fig. 1 Applying a magnetic field to MRE. **a** Without a magnetic field, **b** With a magnetic field

Table 4 Experimental data

G′(Pa)	Error	G″(Pa)	Error	Shear deformation (rad)	Error	Loss factor	Error
B = 0 mT							
1832829,120	3%	432364,5888	8%	0,00192032	4%	0,23590011	9%
1284667,776		314776,2816		0,00484875		0,24502544	
1016428,992		249581,6448		0,00973429		0,24554755	
818620,2240		220044,8448		0,01952960		0,26879967	
680515,9680		189552,7488		0,06856240		0,27854269	
662680,5760		175053,9584		0,07134710		0,25537474	
B= 100 mT							
2146130,496	7%	703627,584	9%	0,00482521	7%	0,32785871	7%
1540270,464		540896,333		0,00517595		0,35116971	
1213273,536		401948,678		0,01934670		0,33129271	
871581,1200		297939,264		0,04831200		0,34183768	
765784,3200		276772,166		0,08939940		0,36142313	
714287,6160		251708,384		0,09605940		0,33639087	
B= 150 mT							
3152750,592	9%	1033410,048	8%	0,00481840	8%	0,327780463	8%
2182366,272		734054,2080		0,00976011		0,336357016	
1626211,392		532352,2368		0,01911990		0,327357341	
1126213,632		348187,2384		0,04810510		0,309166244	
890823,9360		294706,1376		0,08912640		0,390824224	
814648,1280		320599,7184		0,09675360		0,424833385	
B= 300 mT							
2965982,78	7%	724816,704	7%	0,00475383	5%	0,24437657	2%
2580620,54		633542,784		0,00726697		0,24550017	
2313459,07		576327,398		0,00969859		0,24911934	
1703498,11		463652,467		0,01908080		0,27217668	
1316022,91		359488,896		0,04522950		0,27316310	
1223957,82		333615,642		0,04755610		0,27902794	

force whose consequence is to strongly stiffen the chains of particles, which then act as real small fibers. Then, during straining, the elastic stress will exceed the magnetic stress and it will gradually break the fibers in increasingly short elements. On the other hand there is a significant increase in these moduli under the influence of the magnetic field.

The Fig. 3 shows the evolution of the loss factor according to the shear deformation, as this figure shows, the magnetic field plays an important role in the energy dissipation and it notes that the loss factor is growing very strongly with the increase in the magnetic field. However, the angle of loss shows clear differences (Fig. 3): The fraction of the dissipated energy increases with the field and with the creation of the pseudo-fibers formed by the ferromagnetic particles.

The results of the quasi-static tests are illustrated in Fig. 4, a strong magnetorheological effect is observed on the charged composite to 40%, a sign of an interaction between channels. Furthermore this figure gives a detailed overview of the influence of shear deformation on the magneto-dynamic properties, the storage and loss moduli. It is noted that the magnetic field modifies significantly the rheological properties and acts mainly on the shear deformation. In addition, the decrease and the relative increase of G' is a little more intense than for the loss modulus G''; or the addition of oil reduces strongly the local stresses and requires more strong deformations to access critical stresses.

The magnetic field despite all small defects (bad aggregates, bad alignments of chains, particulate columns very close…), which then contributes to the interaction between neighboring chains and during the solicitations, there will be as many additional detachments, which is reflected at the microscopic level by the fall accentuated G' and G''.

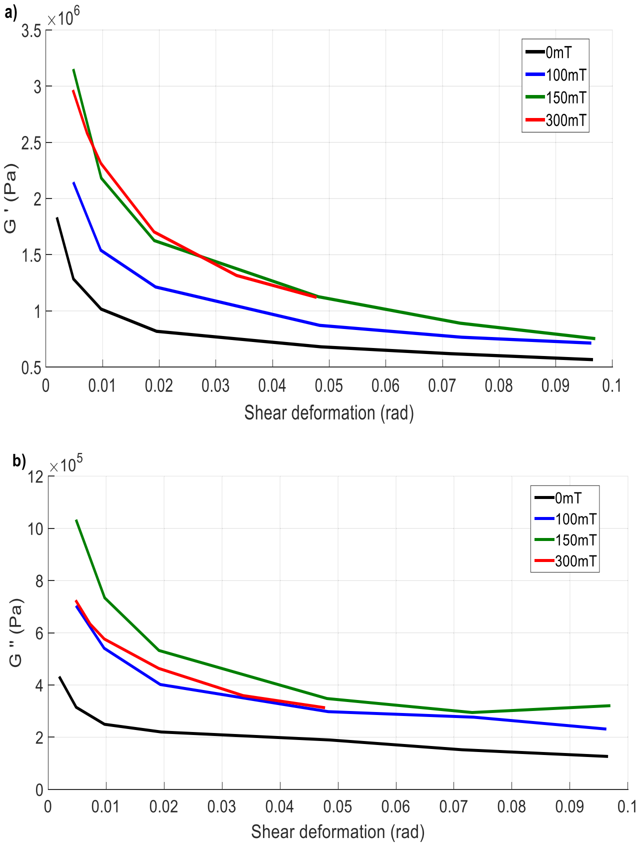

Fig. 2 Variation of rheological properties as a function of shear deformation, **a** Storage modulus, **b** Loss modulus

MREs consist of micrometric particles sensitive to a magnetic field embedded in a polymer matrix. Under the effect of the field the particles are polarized and aligned in the direction of the field. Changes in the spatial distribution of particles by the field are responsible for changes in their mechanical, electrical and optical properties. The fibrillar structure makes the MRE materials very interesting in several applications, they are conductors even at low particle charge rates and are also more sensitive to pressure.

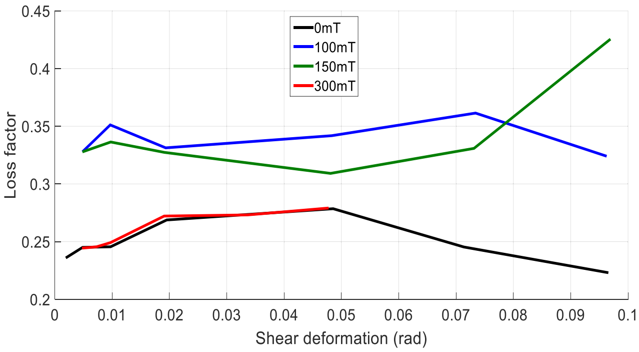

Fig. 3 Variation of loss factor as a function of shear deformation, **a** storage modulus, **b** loss modulus

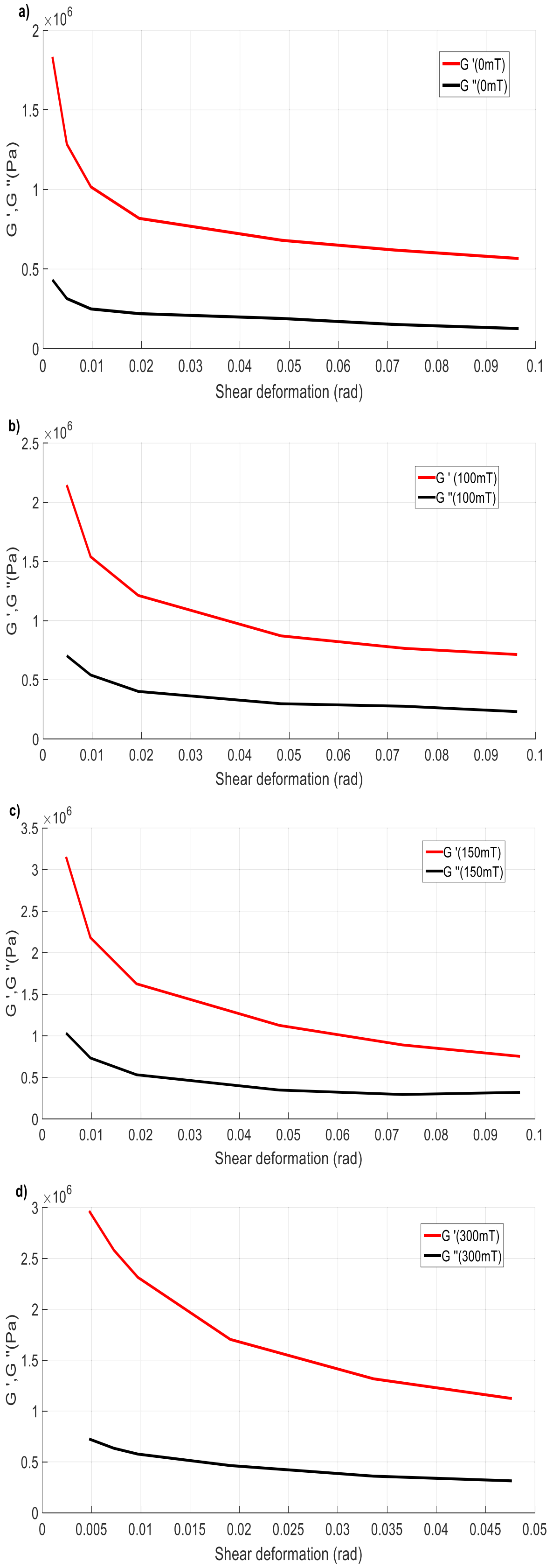

Fig. 4 Variation of rheological properties according to shear deformation for different magnetic field

Structured chains of magnetic particles dispersed in a liquid phase still elastomer creates a microstructure permitting active control of the composite properties via a magnetic field; the increase in stress in the presence of the field is limited by the quality of the microstructure and the ruptures in the chains during the solicitations.

4 Conclusions

The magnetorheological elastomer charged with 40% of iron particles is prepared under different magnetic fields (0mT, 100mT, 150mT and 300mT); the microstructures are greatly affected by the magnetic field intensity during the preparation. The MRE viscoelastic properties are also tested by a mechanical–magnetic coupling dynamic mechanical analyzer (DMA).

The realization of a structured composite elastomer can not be done under any conditions. First, the elastomer must have good mechanical properties but also a low viscosity before crosslinking to facilitate the dispersion and structuring of the charges. Therefore, the conditions of preparation play an important role in the rheological properties of the composite elastomer; the conclusions drawn are given below:

The quantity of iron particles plays a significant role in improving MRE performances, but a big increment of iron particles quantity leads to the decrement of tensile strength and angle tear strength of the MRE.

The interparticle attractive force generated by the magnetic field approximates the particle and modifies the resistivity of the elastomer composite.

Rheological properties increase with the applied magnetic field intensities during testing.

The rheological properties of the elastomer also depend on the arrangement of their particles. The application of magnetic field leads to an important increase in elastic modulus.

The results show a non-linear change in the rheological properties according to the variation of the magnetic field intensity, it is due to the magneto-rheological effect.

An essential advantage of this type of elastomer is to develop shock absorbers capable of damping vibrations in a wide range of frequencies.

Acknowledgments Thanks to Laboratory of Condensed Matter Physics (LPMC) University of Nice - Sophia Antipolis - France, for providing various supports for this study. We are also grateful to Messrs George Bossis Research Director Emeritus and Dr. Kuzhir Pavel of LPMC, for their help.

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