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RESEARCH ARTICLE

Magnetic saturation of poly(dimethyl-siloxane) matrix composites based magnetorheological elastomers

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Abstract

The most common used parameters in applications of magnetorheological elastomers are characteristic data of magnetization curve. Vibrating sample magnetometer (VSM) is a unique technique to record the magnetization curve of small samples. From measurement data the further calculations have to be done for determination of the saturation magnetisation as among other factors some materials can not be saturated with this VSM, but the saturation magnetization can be determine as the fitted curve's parameter. It was found, that the saturation magnetisation depends on filling factor (as rate of components). To get composite saturation magnetisation components saturation magnetisation in proportion of their weight percentage should summing-up. The knowledge of error limits presents a usable tool for further examination.

Keywords

saturation magnetisation · magnetic materials · material testing · VSM

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1 Introduction

Magnetorheological elastomers (MREs) form a subgroup within those materials which are capable of changing both their mechanical properties and their geometry under the influence of the external magnetic field. Consequently, the characteristics of magnetization curve is crucial in the application of MREs. Main field of the application of magnetorheological elastomers is actuator technics. The commonly used parameters in the application are associated with the details of the shape of the magnetization curves. In order to determine the saturation magnetisation we applied vibrating sample magnetometer (VSM) technique in poly(dimethyl-siloxane) matrix based hyperelastic composites. On the bases of experimental data an approximate relation between the saturation magnetisation of two component composites and saturation magnetisation of components and composite filling factors will be suggested.

2 Sample preparation and characterisation

We produced composites of 30 and 75 mass percent respectively using poly(dimethyl-siloxane) as base and one of the 12 components indicated in Table 1. The average grain size and grain-size distribution is shown on Fig. 1. Depending on the used powders type, the geometry of particles do exhibit remarkable differences ranging from sphere (BHS, BHQ) to splint-shaped (FM1. FM2). Composites obtain their hyper elasticity by matrix material while their magnetic features are given by filling materials. This way of composites creation give us an opportunity to develop special constructions [1].

The composite samples were produced by mixing the poly(dimethyl-siloxane) matrix and various kinds of filler materials. The ratio of filler to matrix components is changed within 30-75 mass percent. In spite of high mass percent of filler components the whole composites exhibit elastic behaviour in most cases. The composition as well as the samples are collected in Table 1. The sample code is created as follows: first 3 characters show the filling material type, the following 2 characters are the nominal value of filling factor p, while the last character indicate geometrical type of cross-linked polymer. Here we would like to present an example for better understanding of

Tab. 1. Composition and mass fraction of filler materials

	Fe	O	Ba	B	C	N	Si	Nb	Cu	Zn	Ti	Mn	comment
ALD	100												–
BAF	62,10	32,6	5,30										$BaO{\cdot}6Fe_2\,O_3$
BHQ	> 95	$< 0,5$			$< 0,9$	$< 0,9$							carbonyl iron
BHS	$> 97,8$	$< 0,5$			< 1	< 1							carbonyl iron
EFI EDU	> 98				$< 0,06$		$< 0,12$					$< 0,2$	–
FE3	56,96	38,68					2,58					1,78	$Fe^{2+}Fe_2^{3+}\,O_4$
FM1 FM2	73,5			9			13,5	3	1				Fine-met
M2F	42,51	36,71								8,12		12,66	pro-Ferrite
M2S	44,18	34,86								7,56	0,33	13,06	pro-Ferrite, sintered
P03	54,86	37,71	4,96				2,46						$BaFe_{12}\,O_{19}$

sample codes: ALD75T is a specimen created from ALD filler material (Table 1) and poly(dimethyl-siloxane) in 75-25 mass ratio. The specimen was prepared from material cross linked in disc-shaped moulding tool. The knowledge of mechanical and

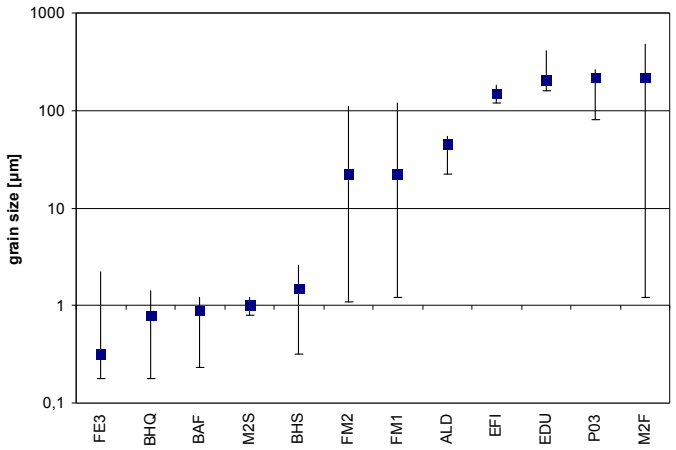

Fig. 1. Grain-size distribution of filler materials

magnetic parameters is needed in the constructions design operation of a given actuator. The proper formulation is required for development of composites with desired magnetic properties namely the relation between component properties and effective properties of composites.

3 Experimental device, data evaluation

Magnetisation measurements were performed by Foner-type (Vibrating Sample [2]) magnetometer in the Research Institute for Technical Physics and Materials Sciences – Hungarian Academic of Sciences. The sketch of measuring principle of VSM equipment is illustrated in Fig. 2.

Foner was the first one who developed the measurement based on flux oscillation induced by vibrating sample located near coils. The ball or cylinder shaped sample is fixed to the end of stick while the opposite end of stick is stimulated by an audio-frequency oscillator. Through the stick the oscillator vibrates the specimen at ∼80Hz and 0.1mm amplitude perpendicularly of line of magnetic force. (Recently a new version of VSM was introduced [3] where specimen oscillates parallel with line of magnetic force. This experimental setup has more advantages than the other one.) The specimen oscillation magnetic field induces alternating voltage in sensing coils. The upper part of the stick is labelled with a little permanent magnet which serves as a reference sample. This reference magnet also induces voltage in reference coils. The voltage of these two coil-pair is compared and their ratio is proportional to magnetic moment of specimen. This method makes the measurement independent of some variables as frequency or amplitude. Measurement setup shown on Fig. 2. is just one among the few previously described by Foner. The equipment has to be calibrated with sample of known saturation magnetisation sample.

The method is universal and extremely sensitive. The magnetic moment sensibility of the equipment is $5 \cdot 10^{-5} {}^{erg}/_{Oe}$ witch matches to $5 \cdot 10^{-9} {}^{emu}/_{g}$ in 10kOe field when related to 1 gram specimen.

This method is not always useable for plotting magnetisation curve in the case of soft magnetic materials, because the demagnetisation force assume high proportion of applied field and therefore the exact effective field inside the specimen is unknown. However this statement does not affect determination of saturation magnetisation as it is independent from the effective field.

4 Measurements

The applied equipment is suitable for the measurements in the range of 0-1.8T magnetic field and 5-900K temperature range respectively. The sensitivity of the equipment is $2 \cdot 10^{-4}$ emu which is generally applicable expect for the thin film specimens.

The measurement data were recorded by continuous sampling. We performed data validation because of some equipment hardware errors. This validation excluded 1.3% of measurement data. The VSM measurement was carried out at 20˚C.

Collection of data was done in four quadrants and mostly in seven sixths as shown on Table 2 and Fig. 3.

5 Consideration about the shape of magnetisation curve

If the sample is composed from small, spatially isolated ferromagnetic particles, being dispersed in non-ferromagnetic media the interaction and demagnetise fields of those particles is

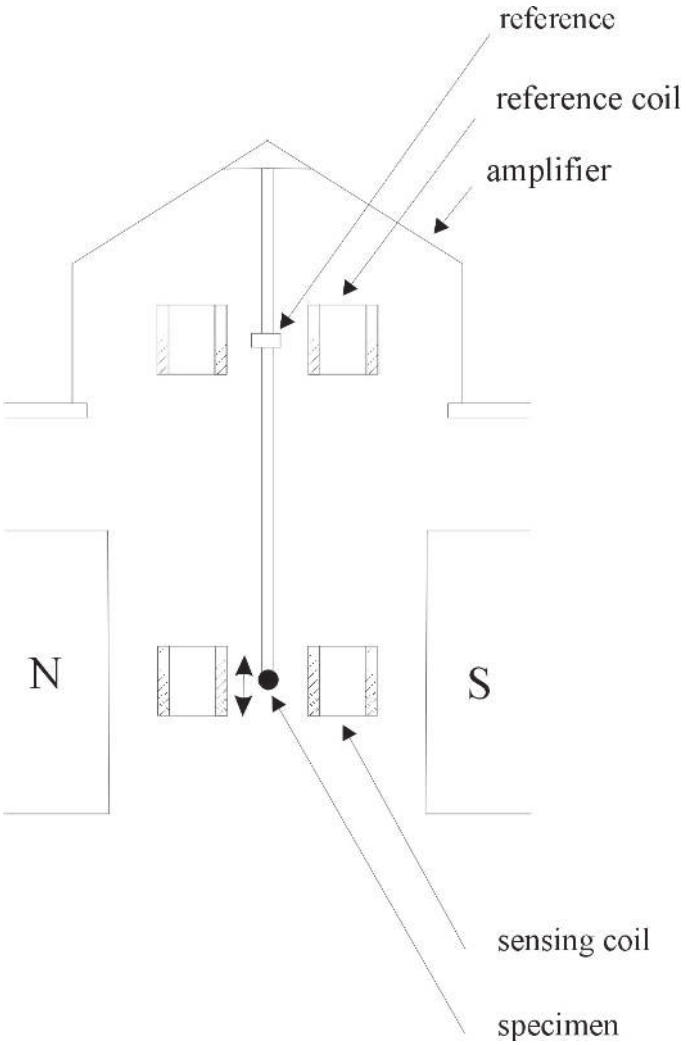

Fig. 2. The working principle and arrangement of VSM

Tab. 2. Measurement signs of VSM measurement

section	$^{dH}/_{dt}$	excitation direction	detector signum
1	+	+	+
2	−	+	+
3	+	−	+
4	+	−	−
5	−	−	−
6	+	+	−
7	+	+	+

remarkable. The macroscopic properties as shape of size distribution and volume fraction of the disperse phase is reflected in $M(H)$ curves. Therefore the microenvironmental (effective) field of particles differs from its macro environmental (applied) fields. To pay respect to self demagnetisation field would prompt us to make complicated correction calculations. Anticipate more difficulties that correction factors are known only for some exact particle geometries [4]. In our case – with exception of BHQ and BHS – it is almost impossible to give an approximate value for the demagnetisation factor.

One possible approach to reach the solution is applying stochastic models for magnetorheological elastomers. This

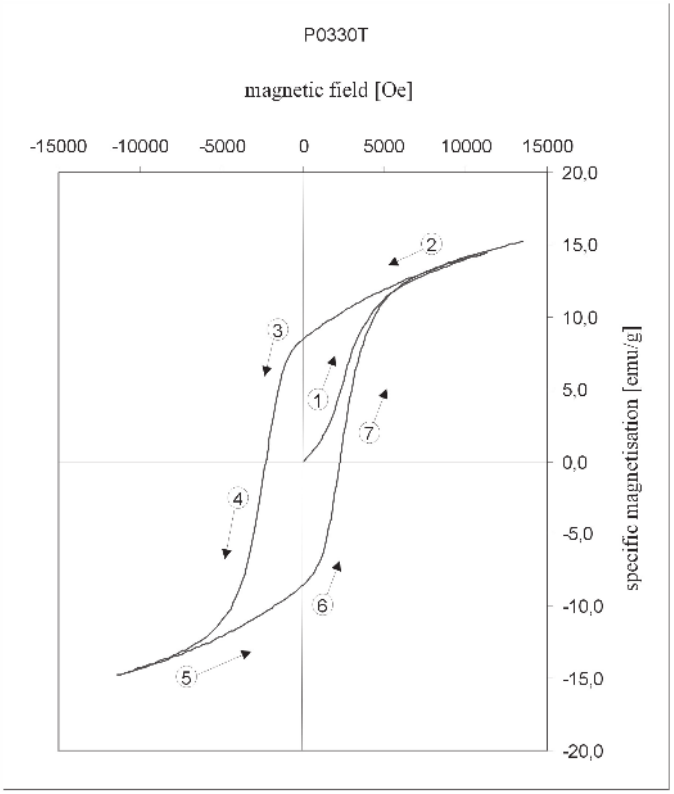

Fig. 3. The details of measuring rum

model is extraordinary complicated and results strictly depend on boundary conditions which are always exposed to high degree of uncertainty. We decided to follow another way, namely to apply a method which has immediate practical use and therefore we used data gained from external field without any correction. It means that data should be considered as $H \equiv H_{ext}$.

From measurement data the further calculations have to be done for determination of the saturation magnetisation as among other factors some materials can not be saturated with this VSM. For example the main field in case of BAF signed specimens would require 17kOe ([5] p578.) for saturation because of anisotropy of crystal-structure.

According to the data obtained from VSM measurements we calculate saturation magnetisation with the still most commonly used series expansion ([5]/9.61) method. Fitting curves are determined by the first three elements of series expansion:

$$M = M_S \left(1 - \frac{A}{H} - \frac{B}{H^2}\right) + \kappa H$$

The fitting carried out for 35 materials (12 materials and 23 composites) in segments located in first quadrant of each VSM measurements respectively. It is brightly visible that segments 1. 2. and 7. (on Fig. 3) converge to the same limit (as saturation) with increasing field. According to findings describe above the fitted curves also result in identical saturation independently of chosen line.

Therefore we decided to chose segment with the optimal geometry giving the preference to the following order: 1, 7, 2. Examples of chosen segments are shown on Fig. 4.

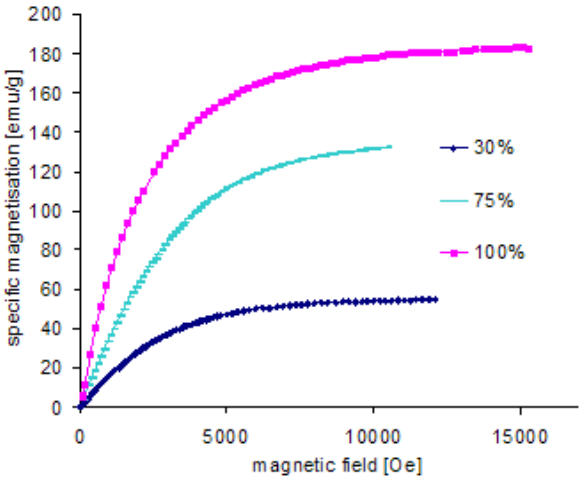

Fig. 4. Chosen segments of VSM measurement in case ALD

To suit the quality criterion of curve fitting it is practical and useful to execute the fitting on localised parts of chosen segments. For the fitting we used OriginPro7.5 program. According to our experience higher importance should be given to initial parameters than to their marginal limitations if we want to emerge the efficiency of stepwise approximation.

During the curve fitting our aim was to reach the $R^2 > 0,998$ (coefficient of determination) value. This was reachable mostly by 1000 iteration steps and in the bigger part of the cases $R^2 > 0,9997$ was fulfilled.

As an example Fig. 5 shows curve fitting of a low filling factor composite. When results are demonstrated by each component

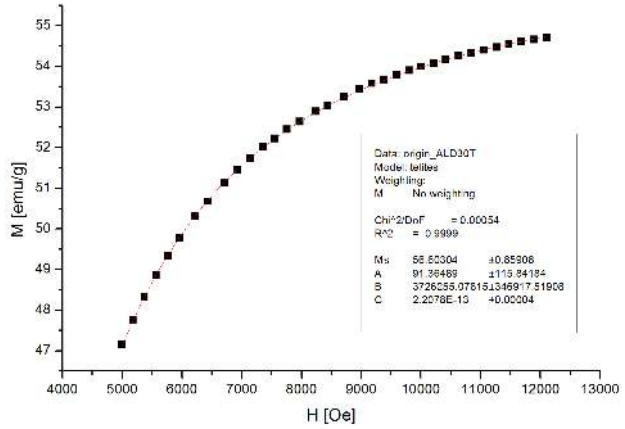

Fig. 5. Curve fitting and its parameters in case of ALD, p=30%

we get the correlation of M_S saturation magnetisation and p filling factor.

The nine interpretable components display the saturation magnetisation values in case of 0, 30, 75 and 100% filling factor. On Fig. 6, the previously multiple date lines (P0330T, ALD75T) are reduced by averaging.

6 Conclusion

Our data suggest that saturation magnetisation primary depends on filling factor (as ratio of components). To get composite saturation magnetisation we should summing-up components saturation magnetisation in proportion of their weight percentage. A composite made of A matrix and B filler material with p filling factor results in saturation magnetisation:

$M_{SC} = pM_{SA} + (1-p)M_{SB}$, that is

$$M_{SC} = M_{SB} + p\left(M_{SA} - M_{SB}\right),$$

where M_{SA} and M_{SB} correspond to materials saturation magnetisation.

The measured (M_{SE}) values differ from calculated values. The absolute difference is: $\Delta = M_{SC} - M_{SE}$, while the relative difference is:

$$\delta = \frac{\Delta}{M_{SE}}.$$

If we reveal absolute values of relative difference $\left(|\delta| = \left|\frac{\Delta}{M_{SE}}\right|\right)$ in each composite than the following values will be obtained, as shown in Table 3.

Tab. 3. Absolute values of relative difference of saturation magnetisation

p [%]	0	30	75	100
filling material	absolute values of relative difference			
M2F	0	0,012	0,069	0
P03	0	0,006	0,036	0
BAF	0	0,240	0,105	0
FE3	0	0,086		0
M2S	0		0,120	0
FM1	0	0,029	0,153	0
FM2	0	0,079	0,122	0
BHS	0	0,032	0,001	0
EFI	0	0,020	0,055	0
BHQ	0	0,099	0,078	0
ALD	0	0,012	0,004	0

The mean value of absolute values of relative difference is 7.8%. Extremely high (more than 10%) error values were detected only in the indicated cases.

In the case of produced and measured composites the relative difference might be even higher but it never exceeds 25 percent. In the background of these differences we established two groups of possible reasons.

- Errors, as
  - the real content of composite is different than the nominal one,
  - chemical and physical parameters of filling materials are modified during the manufacturing process,
  - VSM measurement errors,
  - curve fitting errors,
- neglected interrelations, most frequently
  - interbedding conditions,

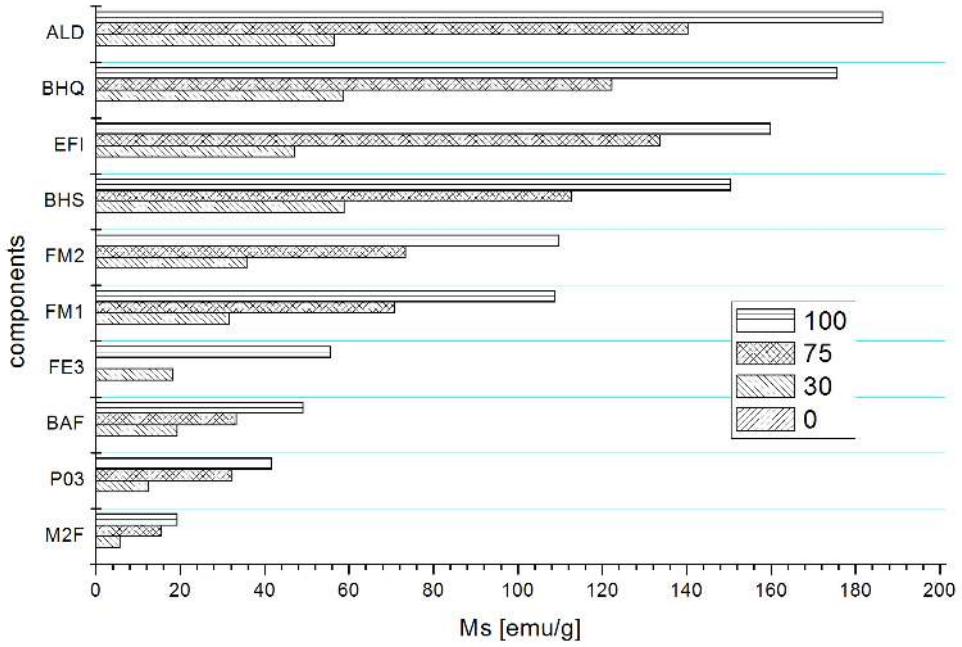

Fig. 6. Filling factor dependents of saturation magnetisation

– effect of macroscopic anisotropy,
– allotrope anamorphism during the production of composite.

Regardless of these errors, as previously written, it is possible to identify composites in a 25% wide range.

7 Case study

The 25% error limit gives an opportunity for band-identification. The measured saturation magnetisation of composite with unknown filling factor but known filler material (M2S) is: $M_{SE} = 53,55$ (all units are given in emu/g). According to saturation magnetisation ($M_{SE\,SIL} \approx 0$, $M_{SE\,M2S} = 80,57$) of components, the area shown on Fig. 7. can be determined with following lines: $M_{S\max} = 1,25 M_{SE} p$ and $M_{S\min} = 0,75 M_{SE} p$. The measured $M_{SE} = 53,55$ value of specimen of interest makes intersections with error lines in points $p_1 = 53,55\%$ and $p_2 = 89,26\%$. The mean value of these parameters is $p = 71,41\%$. Therefore the nominal mass percentage is 70%. The error of 70% filling factor sample calculated ($M_{SC} = 56,40$) and measured saturation magnetisation is $\delta = 4,56\%$, which stays below the average error level.

8 Discussion

Our results suggest a new formula for development of desired saturation magnetisation composites which can be easily applied in the engineering practice. We can state that saturation magnetisation of composites depends mostly on filling factor (namely of filler materials weight percentage). To get composite saturation magnetisation we should summing-up components

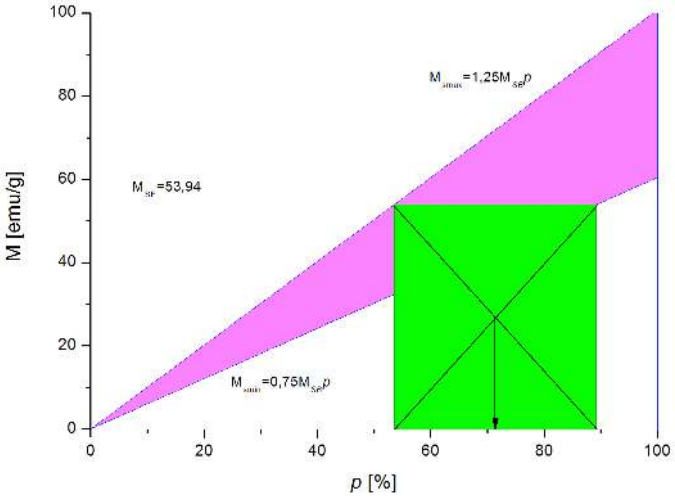

Fig. 7. Identification of M2SxxT

saturation magnetisation in proportion of their weight percentage.

There is a notable difference in our findings when compared to powder metallurgy or two or multiphase solid solution systems. Our formula stays linear even in the case macroscopic filler material particles as it is demonstrated by our experimental setup (Fig. 6 versus Fig. 1). We can also reveal that the geometry of particles has no influence on our results when applied in this grain-size.

The produced composites saturation magnetisation stays under 25% when compared to the calculated value, in average it is 8% difference during the process engineering. The knowledge of error limits presents a usable tool for further examination.

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