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Silicone Magnetoelastic Composite

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Summary: This chapter describes research into magnetic silicone composite properties. The main components of this composite are silicone matrix and magnetic fillers (fine iron and ferrite powders). Composites with changing elasticity can be obtained by modifying matrix composition and the content of fillers. These products are “smart materials”, and it can be expected that they will have wide application in electronics and electrical engineering

The different types of silicone elastomers (polycondensation, polyaddition, and radical vulcanization polymers) were tested as matrices for magnetoelastic composites [1, 2]. Magnetoelastic is a new magnetic composite capable of changing its properties under the action of a magnetic field. The investigated materials represent a new type of composite consisting of small magnetic particles dispersed within a highly elastic polymeric matrix. They are named “smart materials” [3]. These materials are unique in their capacity to increase elongation [3 – 5] and elasticity [6, 7] in a magnetic field by up to 100%.

It was found that the silicone polyaddition polymers are more suitable as a matrix because of the absence of by-products from the process of vulcanization and the formation of ethane bridges (Scheme 1) in the polymer, which significantly strengthens the end product, while high elasticity is retained.

The main object of this research was a study of highly elastic polymer matrix formation processes for magnetelastic composites. An elastic polymer consisting of hydride- and vinyl-containing oligomers was synthesized by hydrolytic copolycondensation of corresponding organochlorosilanes and catalytic polymerization of organocyclosiloxanes with vinyl-containing disiloxanes. The matrix, with tailor-made physical and mechanical performance, was produced by two techniques:

- Varying vinyl and hydride groups ratio in starting components;
- Plasticization of high-strength polymers through introduction of polymethylsiloxane fluid (PMS) into the matrix (Fig. 1).



Scheme 1. Polyaddition polymerization.

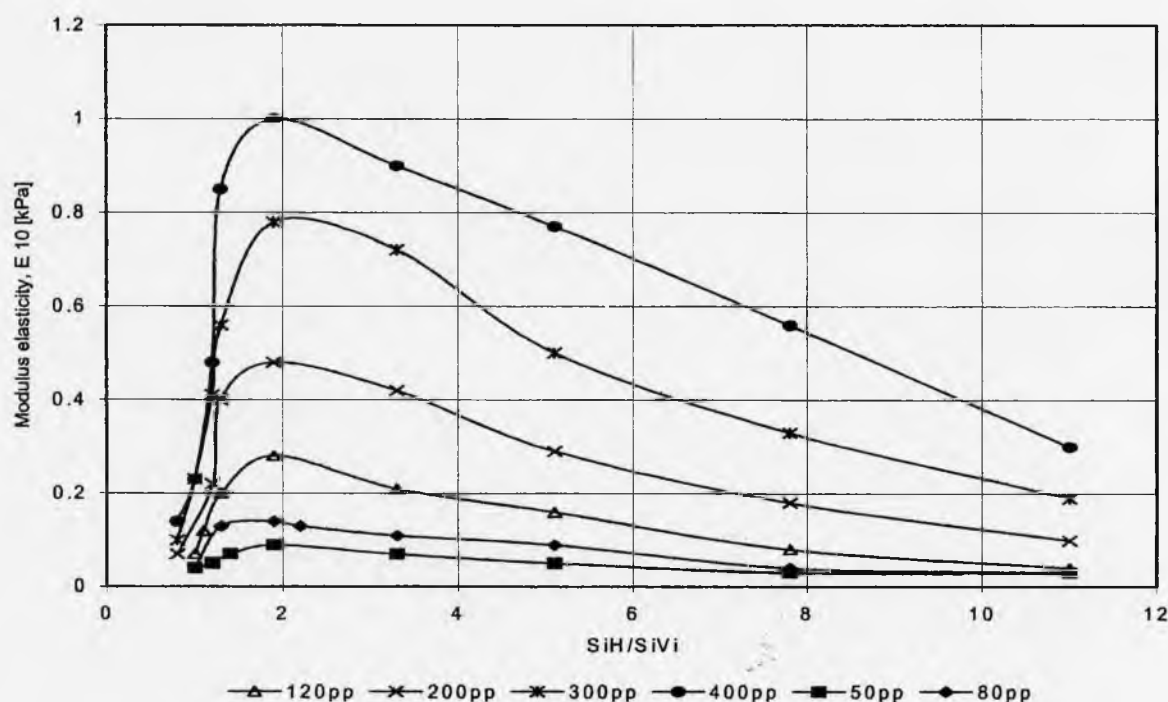


Fig. 1. Modulus of elasticity as a function of ratio SiH/SiVi at different contents of PMS-200.

Magnetoelastic (ME) parameters — elasticity or Young's Modulus (E), strength (P), elongation at failure (L) were evaluated. Strength decrease accompanied by elasticity increase for both synthetic methods of highly elastic matrix was observed, i.e. P/E or E/P ratio was constant for a specified silicone composition. ME was produced by means of mixing silicone matrix with magnet powders. Then the composition was polymerized at high temperature (120 – 150 °C) for 1 – 3 h.

For the purpose of composition optimization of the silicone matrix, two types (I, II) of polyaddition compounds were researched. Special additional agents (QM) were synthesized for increasing tensile strength.

Composition I consists of hydride-containing oligomer ($M_w \sim 800$, content of active hydrogen

0.4 – 0.5%) and oligomer with terminal dimethylvinylsiloxyl units ($M_w \sim 75\,000$).

Composition II consists of the same hydride-containing oligomer and oligomer with terminal trivinylsiloxyl units ($M_w \sim 70\,000$).

The physical mechanical properties for initial and filled materials, with compositions, are presented in Tables 1 and 2.

Table 1. Influence of reinforcing agent on physical–mechanical parameters of composition I.

NN	Content of QM [%]	Initial composition			Filled composition		
		<i>P</i> [kPa]	<i>L</i> [%]	<i>P/L</i>	<i>P</i> [kPa]	<i>L</i> [%]	<i>P/L</i>
1	–	70	179	0.39	400	230	1.7
2	0.2	130	188	0.70	870	282	3.1
3	0.5	270	305	0.90	1050	296	3.5
4	0.8	240	192	1.25	1130	245	4.6
5	1.0	3.90	201	1.95	1270	215	5.9

Table 2. Influence of reinforcing agent on physical–mechanical parameters of composition II.

NN	Content of QM [%]	Initial composition				Filled composition			
		<i>P</i> [kPa]	<i>L</i> [%]	<i>E</i> [kPa]	<i>P/E</i>	<i>P</i> [kPa]	<i>L</i> [%]	<i>E</i> [kPa]	<i>P/E</i>
1	–	450	130	400	1.1	1650	244	560	2.9
2	0.2	760	191	370	2.05	2000	223	324	6.1
3	0.5	1130	239	280	4.03	1550	205	397	3.9
4	0.8	870	224	210	4.14	1130	165	306	3.7
5	1.0	590	210	150	3.9	1030	193	170	6.1

It is seen from Tables 1 and 2 that the introduction of QM increases the tensile strength of polymer matrix and ME while preserving the high elongation (parameter *P/L* increases too). It is very interesting that the introduction of magnetic filler increases the composite tensile strength threefold with some increasing of elongation.

As shown in Tables 1 and 2, other conditions being equal, compositions of series II have higher tensile strength than analogous compositions of series I. This is explained by additional ethane bridges in the polymer net thanks to three terminal vinyl groups.

As seen from Fig. 1 and Table 3, the introduction of plasticizer significantly increases the elongation and decreases the elasticity and tensile strength.

The silicone matrix used for composite production was most favorable, as it allowed simple modifying adjustment of composite viscous and elastic properties according to composition, temperature, and time of polymerization. The presence of filler in the cured composition may

influence mechanical properties in two ways. On the one hand, filled composition strength outperforms the initial strength; on the other hand, active filler surface may partly inhibit polymerization, up to its complete blocking. All samples (Table 3) were prepared on the base composition III (compound SIEL 159 – 254 is the trade mark). So, Fe^{2+} ions can exist on the iron metallic filler and magnetite surface. The initial surface is oxidized and covered by oxide film in which Fe is fully oxidized and is in the Fe^{3+} condition. However, along with the increase of temperature up to 150 °C, exchange processes begin in the filler. The surface oxygen of the oxide film diffuses into filler particles, and Fe^{2+} ions are formed on the surface. These ions are very active inhibitors of polyaddition reaction. The studies showed that the rate of polymerization was decreasing when the filler concentration was increasing; at the same time, the elasticity was decreasing. If the magnetic powder was fresh or old powder was ground and introduced into the polymer, the process of polymerization was practically blocked. Polymerization was not observed at high filling of polymer (~10 vol%).

The study of the influence of plasticizer on physical-mechanical properties of ME is presented in Table 3. Introduction of plasticizer causes elongation increase (up to 1100 %) and reduction of elasticity (up to 1.7 kPa) at the same time.

Table 3. Influence of filler and plasticizer concentration on physical mechanical parameters of composition III.

NN	Filler [%]	Content of plasticizer in polymer [%]	<i>P</i> [kPa]	<i>L</i> [%]	<i>E</i> [kPa]	<i>P/E</i>
1	50	0	1650	–	400	4.1
2	60	0	1720	–	510	3.3
3	70	0	1300	–	220	5.9
4	60	50	28	1130	1.76	16*
5	75	50	173	309	9.6	18*
6	60	0	1800	200	540	3.3
7	60	50	370	410	31	12
8	60	75	53	550	5.5	9.6

* In this case the hydride-containing oligomer was added at the rate of 15 wt%, and the stoichiometric quantity is 10 wt% (Scheme 1, reaction 1). As a result, ratio *P/E* was increased.

Our investigations showed that composite elasticity increase in a magnetic field. The increase in elasticity in a magnetic field is higher when the starting composite elasticity is lower. ME elasticity varied within the range 1 – 500 kPa. In Fig. 2, the dependence of elasticity on magnetic field is shown. For samples with initial elasticity 2 kPa placed in a magnetic field (the value of the magnetic field is 4 kOe), elasticity increases by up to 800%. When initial composite elasticity increases up to 200 kPa, no elasticity increase in the magnetic field occurs.

Detailed study of ME properties demonstrated that in the case of ME synthesis in the magnetic field, significant anisotropy of ME elasticity was observed. Elasticity measured for an ME sample

along the magnetic filler orientation axis is a few times higher than the sample elasticity in the perpendicular direction (Fig. 3).

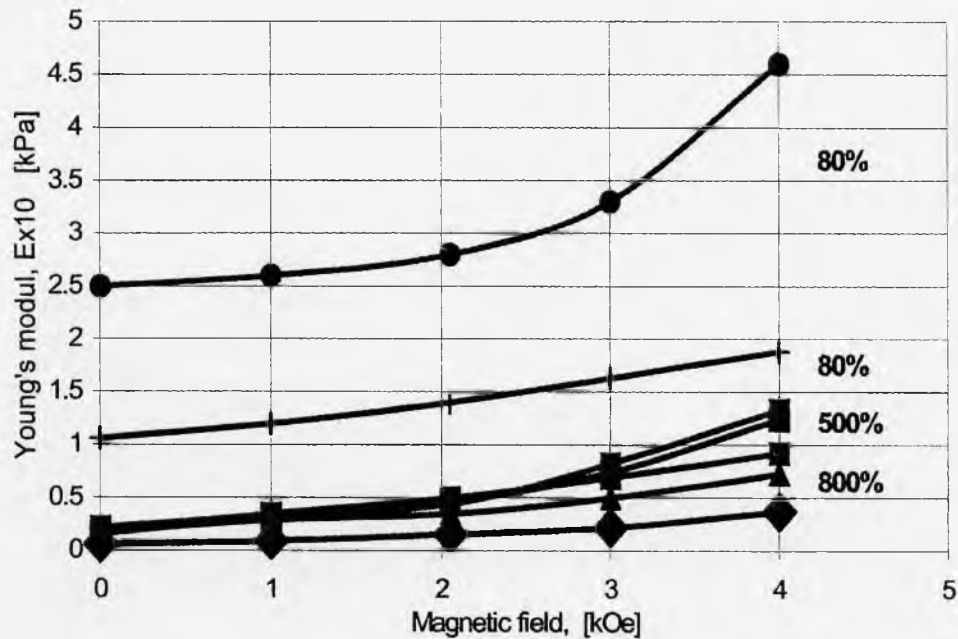


Fig. 2. Dependence magnetoelastic elasticity on size of magnetic field. The percent (%) is ratio of elasticity in magnetic field to initial elasticity.

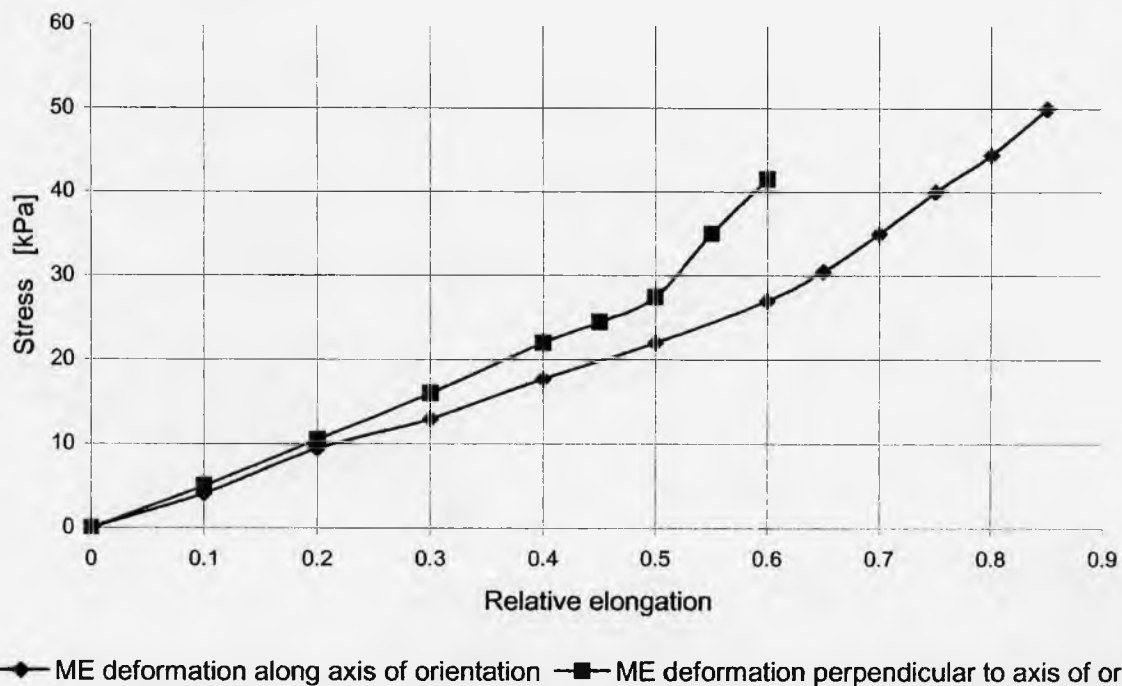


Fig. 3. Dependence of stress in a sample on its relative deformation.

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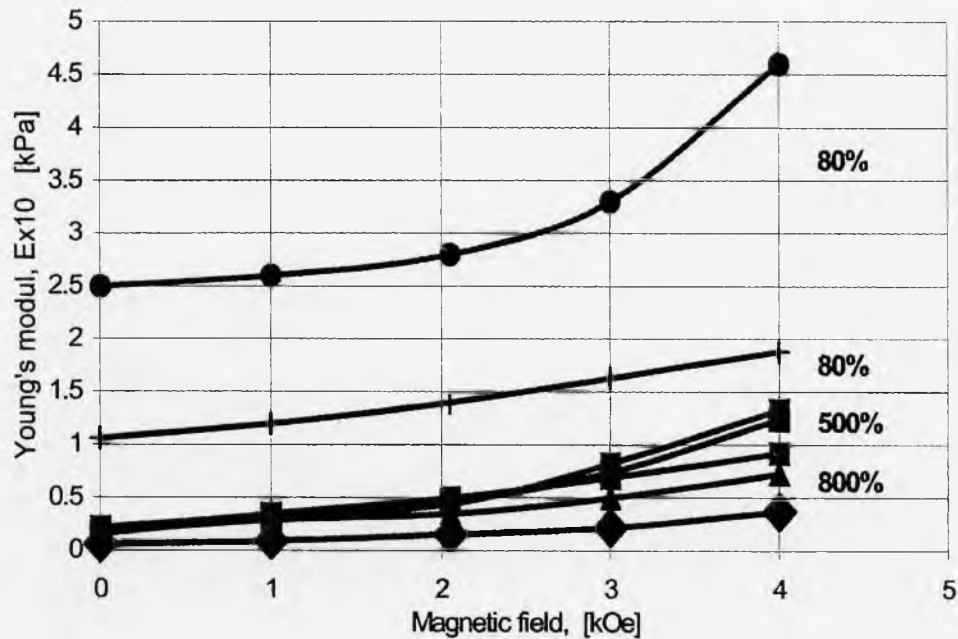


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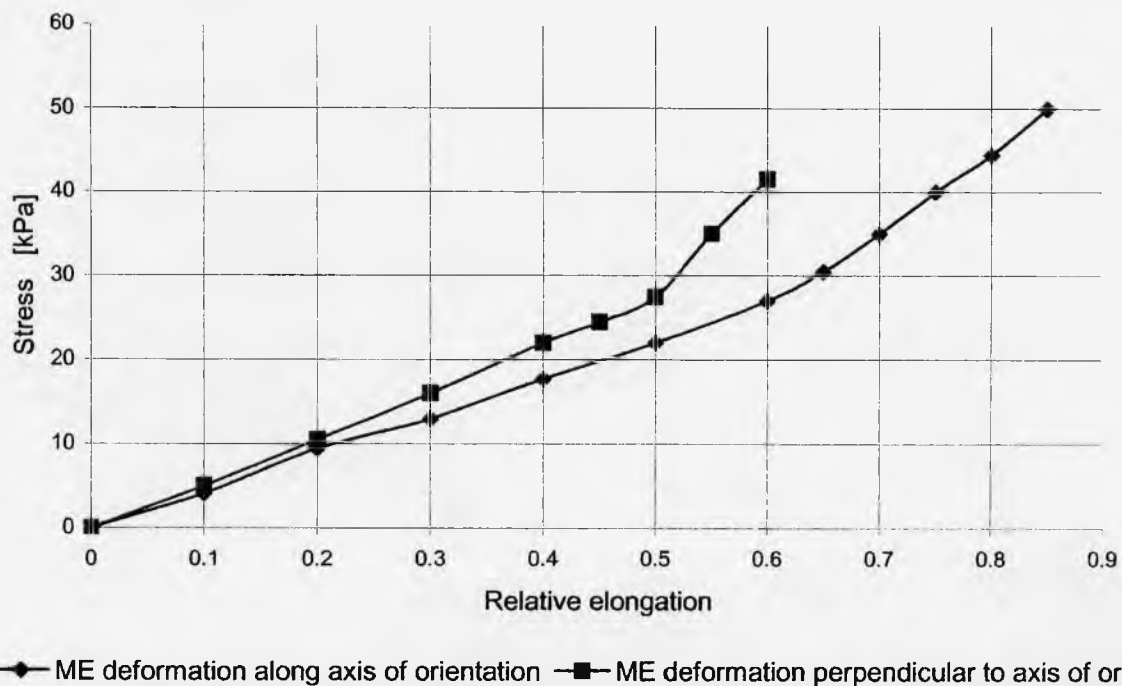


Fig. 3. Dependence of stress in a sample on its relative deformation.

Conclusion

A polymeric matrix based on a silicone polyaddition composition is promising for the creation of magnetic control material. This type of polymer allows us to regulate the elastic properties of the ME in the range 1 – 500 kPa and elongation up to 1100%. In the magnetic field the elasticity increased by 800 %.

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