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Magneto-piezoresistance in Magnetorheological elastomers for magnetic induction gradient or position sensors

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ABSTRACT

Composite material constituted by Fe micro-particles homogeneously dispersed in a silicone matrix, at a volume concentration slightly above the percolation threshold but separated by a thin silicone layer, was produced. The particle magnetic softness and their average size, have been properly improved with respect to previous investigations in order to maximize the piezo-resistive and the piezo-magnetic effects. The optimal combination of magneto-elasticity and piezo-resistivity enables to achieve a record value of magneto-piezo-resistivity sensitivity. An analytical model is proposed to simulate the theoretically expected behavior of electric resistance vs. the applied induction field gradient, so to predict the magneto-piezoresistive response and explain the obtained material tailoring. The experimental results have been in good agreement with the theoretically predicted behaviors, so validating the employed model and the interpretation of the phenomenon. A simple basic application in position sensing is also reported. The analytical model presented in this paper has demonstrated its potentiality to project further improvements, while the experimental results allow for different innovative applications.

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1. Introduction

Magnetorheological elastomers (MREs) made by magnetic microparticles (MPs) homogeneously dispersed in an elastic polymer matrix have been largely investigated [1–9]. These composite microstructures exhibit several interesting properties due to the coupling of electrical, magnetic and elastic properties: piezoresistivity [10–12], magnetoresistivity [13], magneto-piezoresistivity [14].

In particular, a strain can produce electrical resistivity changes depending on the filling particles concentration and the peculiar particle aligning [15–18]. The application of a magnetic field, during MREs production, results on the MPs orientation in the elastic matrix, and it can change the magnetic interaction with the nearest particles, influencing the elastomechanical character of the composite [19,20]. Several investigations have shown that magnetoresistivity is related to both elastomagnetic strain of material matrix and intrinsic magnetostriction of MPs [13,21]. Also thermoresistance response due to thermal-induced strain has been evidenced [13]. Moreover, particle shape and distribution have noteworthy influence on both piezoresistivity and magnetoresistivity in MREs [22].

Recently, our research group has investigated the conditions in which giant piezoresistance appears and has shown that the strain produced by a magnetic induction gradient is able to produce a peculiar type of magnetoresistance due to the elastomagnetic deformation (magneto-piezo-resistivity) [14].

The listed effects in MREs can be used to develop magnetic field sensors [23], energy convertors [24], pressure sensors [25], flexible electronics devices [15] and tunable vibration absorbers [19].

In this paper we show how the optimization of the filling particle magnetic properties, as well as their size, can enhance magneto-piezoresistivity and prospect the use proper MREs as the sensing core of competitive magnetic induction gradient detectors.

2. Material optimization and experimental details

The investigated MRE was made by magnetic MPs homogeneously dispersed into a silicone matrix, in a volume fraction (vol%) of 37%, performing the same procedure extensively described in previous investigations [10,14]. Particular attention was paid in

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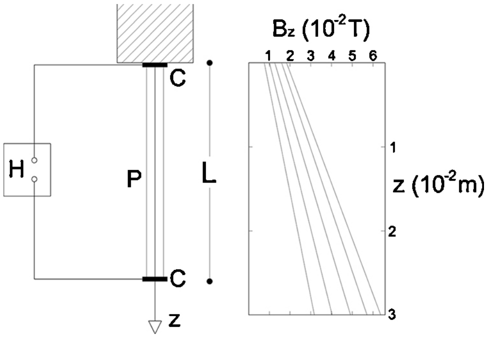

Fig. 1. Scheme of the experimental set-up for magneto-piezoresistance measurements in elastomagnetic composites vertically suspended: P – sample; C – Cu terminations providing the electric contacts; H – resistance meter. A constant magnetic induction gradient was applied along the sample *z* longitudinal axis by means of permanent magnets at a variable reciprocal distance, as described in Ref. [14].

mixing the solid particles inside the liquid silicone component (ESSIL 291, with a viscosity at 25 °C of 43,000 mPa s and a tensile strength of 5 Mpa), before the addition of the cross-linking agent (ESSIL 292 CATALYSEUR, with a viscosity of 4000 mPa s and a tensile strength of 5 Mpa), in order to obtain a thin silicone layer enveloping each particle. For this experimental study bar-shaped samples were prepared (*L* = length = 26 mm; *h* = side of the square cross-section = 2 mm).

The following important changes in the MREs composition and particle average size were performed in comparison with the previously investigated composites [14]:

(i) Iron instead of Nickel was used as magnetic particle material.
(ii) MPs with larger average size were employed (equivalent spherical diameter 7 μm instead of 2.5 μm).

The above listed changes were appropriately tailored in order to improve the sample magneto-piezoresistive response. At first, the substitution of Iron to Nickel enables to obtain higher saturation magnetization values ($M_z = 1.7 \times 10^6$ A/m instead of 0.6×10^6 A/m) at lower magnetizing field. This ensures greater total longitudinal strain $\Delta L/L$ at equal value of E_s (Young Modulus of the used composite material) when a fixed magnetic induction gradient ($\partial B_z/\partial z$) is applied along the sample longitudinal axis *z* as in the scheme of Fig. 1 [14], holding the relation:

$$\varepsilon_z \frac{\Delta L}{L} \equiv \frac{1}{2}(\mathrm{vol\%})\, M_z \frac{\partial B_z}{\partial z} \frac{L}{E_S}. \tag{1}$$

We have already demonstrated [14] that in this type of magneto-elastic composite there is a quasi-linear decrement of electrical resistance logarithm (ln *R*) with longitudinal strain (piezoresistivity). Concerning the effect of MP average size, *D*, on the basis of the Poisson relation $\varepsilon_y = -m\,\varepsilon_z$ (*m* = Poisson Ratio), the local strain, $\varepsilon_g = [(S - S_0)/S_0]$, in the MRE bulk obeys the following relationship [14]

$$\varepsilon_g = -\frac{1}{2}\left(\frac{D}{\eta_0}\right)\varepsilon_z \tag{2}$$

where S_0 is the minimum transversal inter-particle distance, at zero macroscopic strain, *S* is the same distance under a longitudinal macroscopic strain ε_z, and η_0 is the average inter-particle distance at zero macroscopic strain.

Eq. (2) shows that, if the particle size is increased, at a fixed value of the longitudinal strain ε_z, the local strain ε_g will be higher. In turn, if the local strain is increased, the particle proximity is decreased, therefore resulting in a higher decrement of the sample electric resistance.

Thus, the improved MP magnetic softness results in an increase of the longitudinal strain activated by the fixed magnetic induction gradient, applied on the sample. At the same time, the increase of the average particle size should produce, at a fixed longitudinal strain, a larger resistance change. In conclusion, both the operated improvements, (i) and (ii), should cooperate to increase the magneto-piezoresistive effect. This is analytically expressed, due to the model proposed in reference [14], as follows:

$$\frac{R}{R_0} = (1 + \varepsilon_g)e^{\gamma S_0 \varepsilon_g} \tag{3}$$

where R_0 is the resistance at zero strain, and $\gamma = 10.24\sqrt{\phi}\,10^9\ \mathrm{m}^{-1}$ (ϕ = potential barrier for the used metallic element).

Considering Eqs. (1)–(3) one obtains:

$$\frac{R}{R_0} = \left(1 - \frac{1}{4}(\mathrm{vol\%})\, M_z \frac{\partial B_z}{\partial z}\frac{L}{E_S}\frac{D}{\eta_0}\right) e^{\gamma S_0}{}_{-1/4(\mathrm{vol\%})M_z\,\frac{\partial B_z}{\partial z}\,\frac{L}{E_S}\,\frac{D}{\eta_0}} \tag{4}$$

and, if M_z and *D* are increased of a factor 2.8, since both the negative exponent and the subtractive addendum in the brackets have an increment of a factor about 8, a resistance decrement is expected, for the same applied magnetic induction gradient, at fixed values of the dimension *L* and the physical parameters E_S, S_0, η_0 and γ.

3. Theory predictions and experimental results

Considering the parameters characterizing the new MPs, one can affirm that:

(i) Concerning S_0, the use of the same manufacturing process and similar particle roughness, assures a slight difference only from the values obtained in previous experimental investigations.
(ii) SEM analysis has evidenced that the average distance among Fe particles is 1.5 times higher than for the Ni particles used in the previous investigations (this is on-line with the consideration that the Fe particles are bigger than the previously used Ni particles).
(iii) The potential barrier in Fe was expected very similar than in Ni (ϕ = 0.7 eV [14,26]).

On the other side, the Young Modulus determination by stress-strain characteristic of the new Fe MRE samples furnished a value $E_s \approx 7.2 \times 10^5$ Pa, slightly higher than 6.9×10^5 Pa obtained in the previously investigated Ni elastomagnetic samples. This is clearly related to the use of bigger ferromagnetic particles which requires higher stress to have the same strain. Therefore, since the product $D{\cdot}M_z/\eta_0 E_s$ is increased, a higher magneto-piezoresistive response is qualitatively expected in the optimized composite material (higher decrease of *R* with ε_z and, consequently, with magnetic induction longitudinal gradient (Eq. (1)).

In the following, the improved elastomagnetic samples will be indicated as Fe MREs while the already investigated elastomagnetic samples will be reported as Ni MREs.

To clarify the modeling expectation, the comparison between the theoretic behaviors of *R* vs. $\partial B_z/\partial z$ obtained by Eq. (4) is reported in Fig. 2: curve (a) and (b) represents the predicted response of Fe MRE and Ni MRE, respectively.

It is observed that:

(i) The value of the initial resistance R_0 is higher in Ni MREs than in Fe MREs ($9.5 \times 10^{11}\ \Omega$ instead of $6 \times 10^{11}\ \Omega$) and this can be attributed to the larger number of silicone barriers in the old composite due to the lower MP average size.

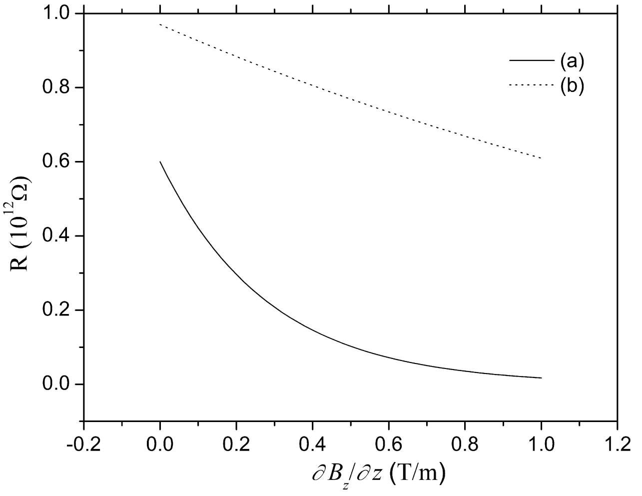

Fig. 2. Comparison between the resistance behaviors vs. the applied magnetic induction gradient, as simulated by using Eq. (4), for the Fe MRE samples (a), and the previously used Ni MRE samples (b).

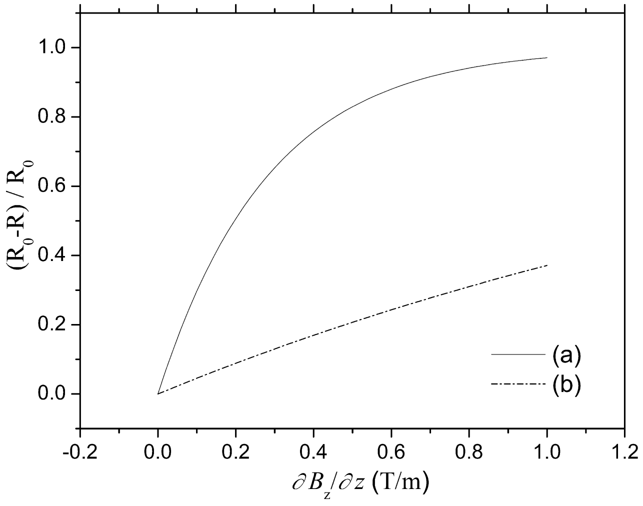

Fig. 3. Simulated behaviors of the magneto-piezoresistance ratio vs. the applied magnetic induction gradient for the Fe MRE samples (a), and the previously used Ni MRE samples (b).

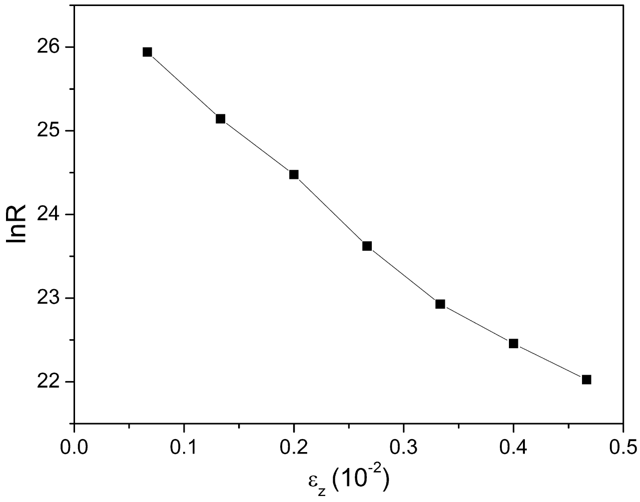

Fig. 4. Experimental curve of ln R vs. longitudinal strain (ε_z) in Fe MREs. Average values of ln R obtained by reiterated measurements are reported. The error bars are too small to be visible. The line is a guide to the eye.

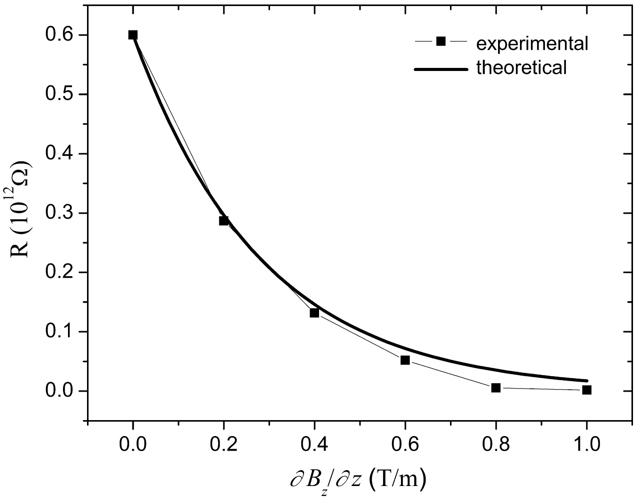

Fig. 5. Comparison between the theoretical behavior of the resistance (Eq. (4)) and the results obtained by averaging experimental values in repeated measurements at fixed values of the applied magnetic induction gradient with steps of 0.2 T/m, in Fe MRE new sample.

(ii) The decrease of R with B gradient is higher in Fe MREs than in Ni MREs, in agreement with the expected optimization discussed above.

In Fig. 3 the comparison between the theoretical behaviors of the magneto-piezoresistance ratio $\mathrm{MPR} = [R_0 - R(\partial B_z/\partial z)]/R_0$ is reported: the large improvement of the new composite material is clearly confirmed.

Therefore, the model simulations, shown in Figs. 2 and 3, confirm the qualitatively predicted expectations.

The direct experimental results on piezoresistive response and magneto-piezoresistance ratio are reported hereinafter. The experimental methodology for these measurements was the same used for the Ni MREs, as detailed in reference [14], thus allowing for an objective comparison.

In Fig. 4, the natural logarithm of the Fe MRE resistance (ln R) as a function of the applied longitudinal strain is reported. These samples have a Fe MPs volume percentage of 37% (±1.5%). This percentage represents the threshold to maintain a sample resistivity comparable to the silicone resistivity. Above this percentage the resistance (of Fe MREs with similar size) decreases, indicating the start of electrical conduction between adjacent MPs.

The piezoresistance sensitivity ($\Delta R/\Delta\varepsilon_z$) is increased to about $3\ 10^5\ \Omega/10^{-4}$ reaching values almost ten times higher than in Ni MREs, at the same MPs percentage [14].

The dependence of resistance versus the applied magnetic induction longitudinal gradient is illustrated in Fig. 5: a giant magneto-piezo-resistance sensitivity ($\Delta R/(\Delta(\partial B_z/\partial z)) \geq 0.5 \cdot 10^{12}\ \Omega\mathrm{m/T}$) has been shown. The good agreement between the theoretical prediction (Eq. (4)) and the experimental measurement, demonstrates that our model is effective to describe the physical phenomenon of coupling among strain, due to magnetic induction gradient, and conductivity, in the new investigated MREs. The obtained improvement of performance offers more incentives for possible applications of the new Fe MREs as sensing core of contactless detection of position, or low frequency vibration, both in civil and mechanical or aeronautical engineering components.

A possible example of position (or small displacement) sensor is shown in Fig. 6. The Fe MRE sample (of the same shape and size used for the other experimental measurements reported in this paper) is set opposite to a coaxial cylindrical magnet, which undergoes a 0.05 mm/min displacement by means of a mechanical cursor, with real-time recording of the sensing core resistance.

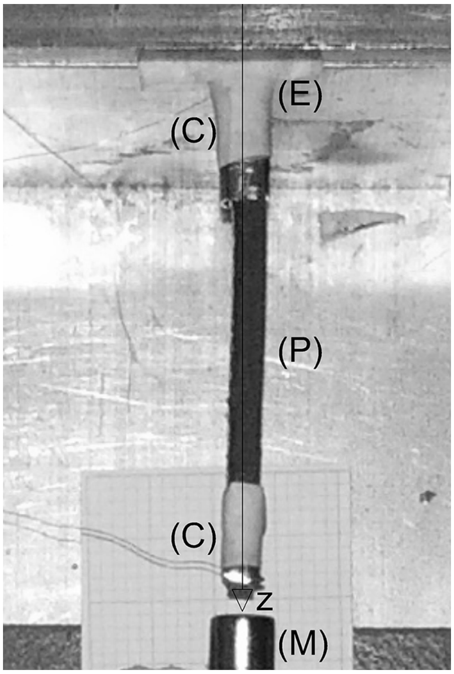

Fig. 6. Prototype of a position (or small displacement) sensor: M – cylindrical permanent magnet; P – Fe MRE sample; C – Contact Cu capillary wires providing electrical contact with the resistance meter; E – clamped extremity.

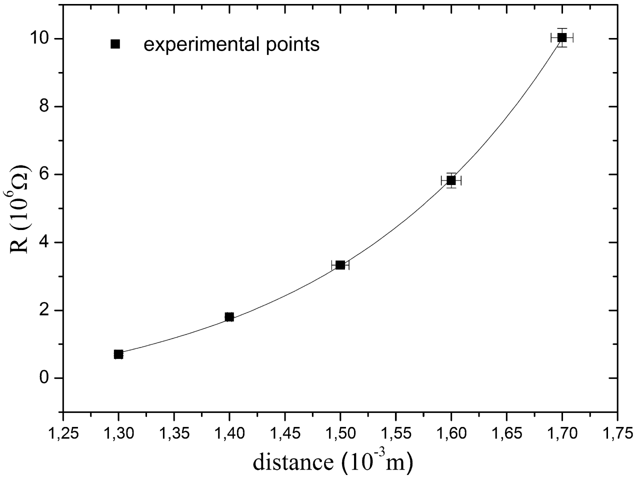

Fig. 7. Calibration curve of a position sensor with a Fe MRE sensing core. Average values of *R* and correspondent standard deviations of reiterated measurements are reported. The line is a guide to the eye.

The dependence of the sensing core resistance on the distance between the sensing core and the magnet surface is represented in Fig. 7. This device can be used to monitor the displacement of any non-magnetic object on which the permanent magnet is set. Taking into account the experimental errors, in the investigated displacement range, an average sensitivity of about 25 kΩ/μm is obtained with an error in position less than 20 μm.

The miniaturization of such a device to micron dimensions, through the use of nanoparticles, may also lead to other useful applications, such as magnetic read heads.

4. Final remarks

The magneto-piezoresistive effect performance in composites of magnetic particles homogeneously dispersed in a silicone matrix was improved in comparison with our previous investigations [14]. The reported experimental results demonstrate that the performed optimization of particle composition and average size has been able to enhance the piezoresistivity and the magneto-piezoresistivity sensitivity. In particular, as already evidenced in Section 3, piezoresistivity increases of a factor 10 and, considering that a magnetic induction field gradient of almost 3 T/m is necessary to produce a *R* percentage change around 60% [14] in the previous Ni MER samples, while 1 T/m induces a *R* percentage change of about 95% in new Fe MERs, an increase of magneto-piezoresistivity of about a factor 3 is obtained (taking also into account the R_0 values difference in the two MER materials reported in Section 3). The performance improvement has been explained by means of a model describing the electron transport resistance versus the magnetically induced longitudinal strain. The response of the new MRE composites demonstrates a good agreement between the model and the experimental results, thus constituting a useful basis for further improvements. The use of the giant resistance change, induced by a moderate gradient of magnetic induction, can be employed in quasi-static displacement sensing devices (mechanical sensors, position detectors) or in sensors of any source producing local magnetic induction gradient (magnetic read heads, provided that a miniaturization to micron dimension is obtained by means of the nanoparticles employ).

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Biographies

Giovanni Ausanio obtained the degree in Physics in 1997 and the PhD degree in Materials Engineering in 2004 at “Federico II” University of Naples. He is currently University researcher. His main research topics are the production, characterization and application of amorphous magnetic materials, elastomagnetic composites, and nanostructures obtained by chemical synthesis and femtosecond laser ablation.

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Enrico Ricciardi was born in Naples, Italy, in 1986. In 2009 he received the B.S. degree in Civil Engineering (with honors) from “Federico II University of Naples” and then, moved to Madrid, at the Polytechnic University, in the frame of the International Study Abroad Plan (Erasmus Program). He received the M.S. degree (with honors) in Hydraulic and Transport Systems Engineering in 2011 from “Federico II” University of Naples. In the same year he took the Cervantes DELE at the Spanish Study Center in Naples. In 2012 He passed the qualifying examinations as an engineer. Winner of a scholarship supplied by Italian Ministry of Education, he is now a PhD Student in Structural Engineering and is working on elastomagnetic material applications in civil engineering, especially in the field of vibrations attenuation.

Luca Lanotte received his Master Degree in Chemical Engineering from the University of Naples “Federico II” in 2009. From 2010 to 2013, he conducted a project in collaboration between the Department of Chemical Engineering, Materials and Industrial Production (University of Naples “Federico II”) and the Laboratoire Interdisciplinaire de Physique (University of Grenoble “Joseph Fourier”) and he is obtaining his Ph.D. in Chemical Engineering from the University of Naples and Physics from the University of Grenoble. His research focuses on rheology and microfluidics of biological fluids, rheo-optics, production of scaffolds by electrospinning, and multifunctional magnetoelastic materials. The results of his brief scientific career are already reported on 5 papers published on international scientific journals, in one patent, and they have been presented in 8 international conferences and meetings.

Luciano Lanotte is full professor of Experimental Physics at the University of Naples “Federico II”, Chief of the Magnetism Group both in the Department of Physics of this University and in the SPIN Institute of the Research National Council, Naples Unit. He published more than 100 papers in the field of new magnetic materials modeling and application, and gave several invited talk on elastomagnetism. In particular, he was: Chairman of the 1st MEA ‘93 “International Conference on Magnetoelastic Effects and Applications” Capri, 24–26 May 1993; Organizer of the Symposium “Magnetostrictive, magnetoelastic, multiferroic, ferromagnetic shape memory materials and application” for Joint European Magnetic Symposia, JEMS 06, San Sebastian (Spain) 26–30 June 2006; Organizer of the Symposium “Magnetic shape memory, magnetoelastic and multifunctional materials” for Joint European Magnetic Symposia, JEMS 12, September 9–14, Parma, Italy. He was awarded as valued reviewer in 2009, for the contribution given to the quality of Sensors and Actuators A.