Modification of the Ultrasonic Properties of Elastomers Loaded with Magnetic Particles by Applying Magnetic Fields During Curing.

I. Agirre Olabide, M. J. Elejabarrieta and M. M. Bou-Ali
Mechanical and Manufacturing Department
Mondragon Goi Eskola Politeknikoa
Mondragon, Spain
Iker.agirreo@alumni.mondragon.edu

M. D. Fariñas and T. E. Gómez Alvarez-Arenas
Sensors and Ultrasonic Technologies Dept. ITEFI,
Spanish National Research Council (CSIC)
28006 Madrid, Spain,
t.gomez@csic.es

***Abstract*— Particle loaded polymers and elastomers have been largely used in different ultrasonic applications like backing materials and matching layers because composite properties (velocity, attenuation, density and impedance) can be easily engineered by changing the filler concentration. Recently, the use of magnetic particles has been theoretically proposed as a means to produce active matching layers whose response can be modified upon the application of magnetic fields. In this paper, we propose to introduce modifications of the composite properties by using magnetic particles and applying magnetic fields during curing to establish well defined patterns in the spatial distribution of the particles within the elastomer that induce material anisotropy that effectively modify ultrasonic properties.**

***Keywords—* Magnetorheological elastomers, ultrasonic properties, isotropic and anisotropic, smart magnetic materials.**

I. Introduction

Magnetorheological elastomers (MRE) are considered smart materials due to the possibility to modify their properties when an external magnetic field is applied. Basically, MRE consist on magnetic particles embedded in a polymeric matrix, where many different polymeric matrices can be used: natural rubbers, silicone rubbers, etc [1-2], with the unique requirement that the matrix has to be paramagnetic. In the ultrasonic field, 0-3 connectivity composite materials have been used as backing and matching layers materials.. More recently, 0-3 connectivity composites made with magnetic particles have been proposed either to change the magnetic properties of the composite [3] or to produce active matching layers whose properties can be changed upon the application of a magnetic field [4].

The most common magnetic material used for the synthesis of MRE are iron micro-particles, which size is between 1 µm and 10 µm [5-6]. However, larger particles until 200 µm and irregular shape particles have also been used [7].

These particles can be randomly distributed within the polymeric matrix or aligned in chains. The alignment of the particles is obtained by the application of an external magnetic field during the so-called pre-structure process [6], due to the interaction between the particles; these samples are called anisotropic MRE to differentiate them from samples produced without any magnetic field where the particles present a random and globally isotropic distribution [7-8].

The aim of this work is to determine and analyze the influence of the particle concentration and the pre-structure process on the ultrasonic properties of the MRE as a first step towards achieving materials whose properties can be either engineered during fabrication or altered after fabrication…

II. Synthesis

The matrix used in this work was a two components silicone rubber. The silicone WACKER Elastosil® M 4644 A and the vulcanizer WACKER Elastosil® M 4644 B, both are mixed in the ratio10:1 respectively.

Carbonyl iron powder particles (average particle size of 1.25±0.55 µm) and spherical shape, supplied by BASF (Germany) were used to make the samples. The samples were produced by mixing the silicone rubber A component and the particles and subjecting the mixture to vacuum cycles to extract the air bubbles generated during the mixing process. Subsequently, the vulcanizer was added and mixed; then, vacuum cycles were applied at the same conditions. The cycle quantity varies with the particle concentration. Once a homogeneous mixture is obtained two different pre-structure conditions were applied to obtain isotropic or anisotropic MRE samples, respectively: under an external magnetic field and without its influence. The thickness of the samples is of 1 mm in all cases.

Anisotropic samples have been pre-structured by a device where neodymium magnets are applied (Fig. 1). Thanks to the ferromagnetic plates, the device guaranties a homogeneous magnetic field. That magnetic field is applied in the thickness direction of the sample [7]. The intensity of the field during the pre-structure is 0.13±0.01T and is measured by a Gaussmeter FH-54.

The present study has been partially supported by ACTIMAT and MAGNETOBUSH (UE2013-09) projects, Research Group program (IT557-10) from the Basque government, and by the MAGNETO (INNPACTO-020000-2010-006), AVISUINT (DPI 2012-36366) and NOVTUL (DPI2011-22438) from the Spanish government.

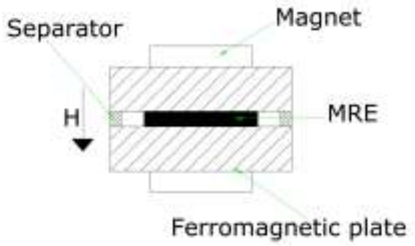

Fig. 1. Sketch of the anisotropic pre-structure device.

Samples with seven different values particle volume concentration were synthesized, 0%, 5%, 10%, 15%, 20%, 25% and 30%, respectively. The maximum concentration is defined by the CPVC [5]. As explained before, two pre-structure conditions were used to obtain either isotropic or anisotropic samples.

III. ULTRASONIC CHARACTERIZATION

Several ultrasonic techniques have been tested to determine the ultrasonic properties of these materials: ultrasonic velocity and attenuation and the variation in these parameters with the frequency. In all cases a through transmission technique was employed, but different coupling methods were tried: water immersion, direct contact and gel coupling and air-coupling. Obtained results were similar, but the air-coupled technique is preferred because it offers a better possibility to study changes in materials properties upon the application of a magnetic field; it is also more convenient to measure shear waves and hence to get shear elastic modulus and Poisson's ratio which will be the next steps of this investigation.

The technique is described in [8]. The magnitude and the phase spectra of the transmission coefficient of a plate-shaped sample are measured at normal incidence and in a frequency band were, at least, one order of thickness resonances of the sample is observed. Effective properties of the material are obtained by solving the inverse problem.

The measurements were performed using a pair of air-coupled wide-band transducers fabricated at *US-Biomat* research group (ITEFI, CSIC) [9], [10]. Transducers were embedded in a U-shaped holder that keeps them aligned and also provides a slot for the correct placement of the sample (see Fig. 2). Transmitter transducer were driven by a 200 V amplitude semicycle of square wave, provided by a conventional pulser (Panametrics 4077), the received signal was analogically amplified (30 dB), with the reception function of the Panametrics 5077 and then digitized by a Tektronix scope (TDS5052). The transmitted signal from transmitter to receiver without any sample between them is shown in Fig. 3 (up). Fig. 3 (down) shows the sensitivity vs frequency calculated as the ratio of the amplitude of the FFT of the electrical signal measured at receiver terminals to the amplitude of the FFT of the electrical signal measured at the transmitter terminals.

Fig. 2. Picture of the transmitter-receiver pair of transducers embedded in the U-shaped holder for air-coupled through transmission measurements. The slot between trnasmitter and receiver is used to place the samples.

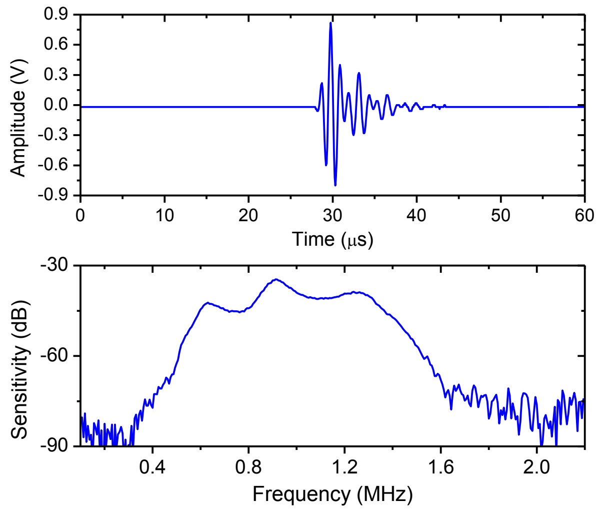

Fig. 3. Up: transmitted signal from transmitter to receiver in the time domain without any sample between them. Down: sensitivity vs frequency

IV. RESULTS

Below are shown the results obtained by the air-coupling technique in the range of 0.34 and 0.95 MHz. First of all, the temporal signal measured by the transducers is analyzed and the quality factor (Q factor) is obtained. Towards this end, the decline of the temporal signal has been adjusted by an exponential function. Finally, the results obtained by the analyzing the amplitude spectrum are shown: longitudinal velocity and the quality factor.

Fig. 4 shows the temporal signal measured by the transducers. But the data used at the present work corresponds to the rectangular window indicated at the figure. The temporal signal corresponds to a single harmonic oscillation under damped system.

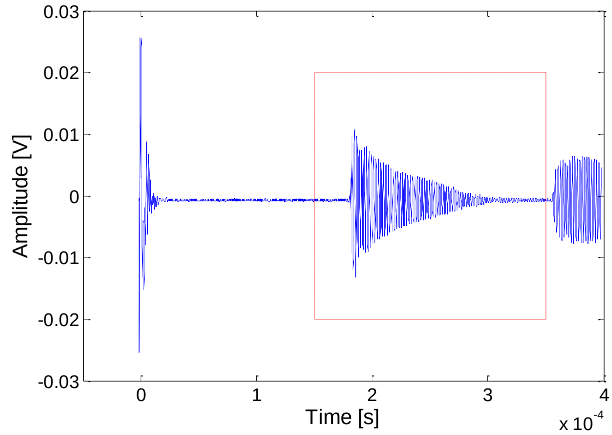

Fig. 4. Temporal signal of the sample without particles.

From the temporal signal the oscillation period, T is defined (Fig. 3). The maximum points of each oscillation are approximated by the least squares method to (1), where the amplitude (A_0) and the quality factor (Q) are defined.

$$A_0 \cdot e^{-\left(\frac{1}{2Q}\omega t\right)} \tag{1}$$

where t is the time and ω is the fundamental natural frequency of the system. That frequency is obtained by (2) and the period (T), for the case of a week damping.

$$\omega \cong \frac{2\pi}{T} \tag{2}$$

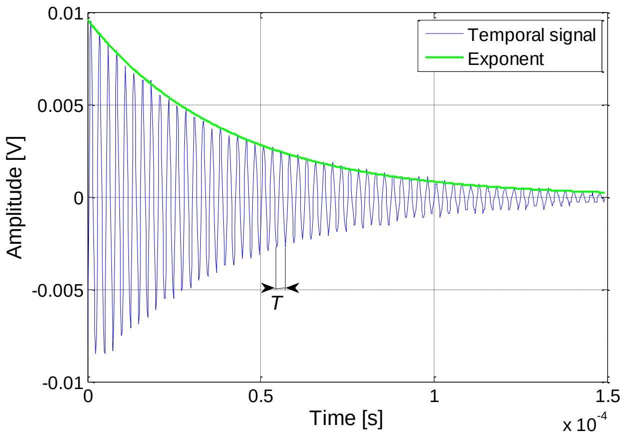

Fig. 5. Temporal signal and the approximation by a exponential function.

The results obtained from the analysis are shown in Fig. 5. The factor decreases with the increment of the particle concentration, which means that the temporal signal attenuation increases with the concentration. Besides, the anisotropic samples has a higher attenuation than the isotropic ones, in other words, the alignment of the particles increases the damping.

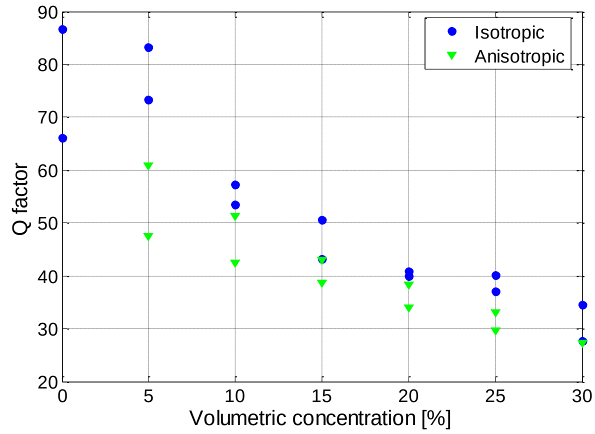

Fig. 6. Quality factor obtained by the temporal signal in function fo the volumteric concentration.

After the analysis of the temporal signal, the Fourier transform has been obtained, and an approximation has been made to obtain different parameters (Fig. 6).

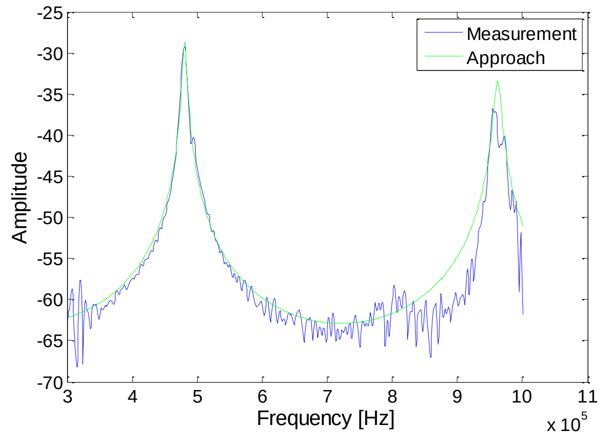

Fig. 7. Spectrum modulus, experimental and the approach.

From the spectrum approach longitudinal velocity is obtained (Fig. 7). Whereas, the velocity decreases with particle concentration, the influence of the pre-structure cannot be observed. This means that the ultrasonic wave has more difficulties with a higher particle concentration.

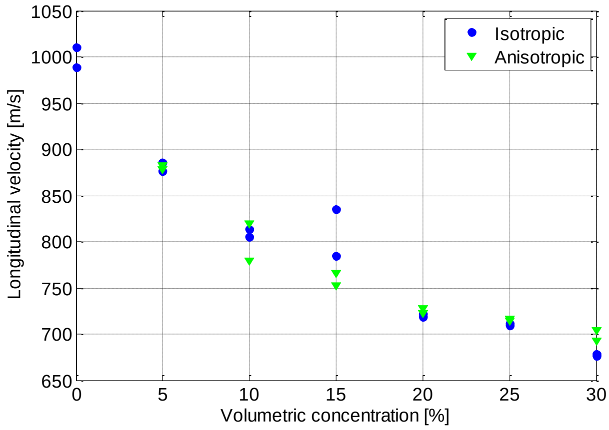

Fig. 8. Longitudinal velocity from the spectrum approach in function of the particle volume fraction.

The quality factor also is obtained from the spectrum approach (Fig. 7). The factor decreases with the increment of the particle concentration and the damping is larger for anisotropic samples.

Those trends are observed using two different techniques, but there is a difference in the value. This is because at the spectrum analysis a calibration is used to eliminate the band effect of the transducers.

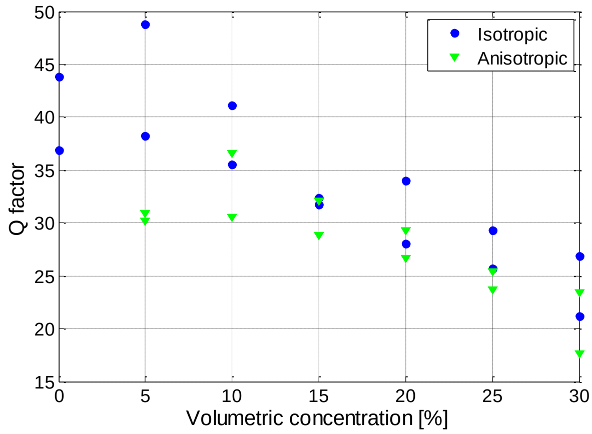

Fig. 9. Quality factor obatained from the spectrum approach at the resonance frequency in function of the volumetric concentration.

V. Conclusions

The increase of the particle concentration from 0 to 30% reduces the longitudinal velocity from 1000 to 700 m/s. This effect is the combined result of the density increase of the composite when the particle concentration increases plus the reduced influence of the particles on the overall elasticity of the composite. This behavior follows the theoretical predictions of Hashin and Shtrikman [11] and the measurements performed before in polymer loaded with non magnetic particles [12] and [13]. However, no effect of the pre-structure obtained with a magnetic field has been observed on the ultrasonic velocity of the longitudinal wave in this frequency range. On the other hand, the quality factor also increases when the particle concentration increases, which means that the attenuation is higher. This larger attenuation is the result of the scattering losses introduced by the particles; In this case, the quality factor depends on the pre-structure process: the anisotropic samples present a higher attenuation than the isotropic samples. Further investigations will focus on the study of the propagation of longitudinal waves in the plate direction and shear waves in the direction normal to the plate. In this case, it is expected that both velocity and attenuation will present different values depending on the direction of the polarization compared with the direction of particle alignment.

References

[1] L. Chen, X. Gong, W. Jiang, J. Yao, H. Deng and W. Li, "Investigation on magnetorheological elastomers based on natural rubber," J.Mater Sci., vol. 42, pp. 5483-5489, 2007.

[2] M. Kallio, "The elastic and damping properties of magnetorheological elastomers," VTT Publ., 2005.

[3] M. Teirikangas, "Advanced 0–3 ceramic polymer composites for high frequency applications." PhD Thesis, Dept. of Electrical Engineering, University of Oulu. 2011

[4] A. J. Mulholland, R. L. O'Leary, N. Ramadas, A. Parr, A. Troge, R. Pethrick, and G. Hayward, "A theoretical analysis of a piezoelectric ultrasound device with an active matching layer," Ultrasonics, vol. 47(1-4), pp. 102–110, 2007.

[5] A. Boczkowska, S. F. Awietjan, T. Wejrzanowski, and K. J. Kurzydowski, "Image analysis of the microstructure of magnetorheological elastomers," J. Mater. Sci., vol. 44, pp. 3135-3140, 2009.

[6] J. Li, X. Gong, Z. Xu, and W. Jiang, "The effect of pre-structure process on magnetorheological elastomer performance," International Journal of Materials Research, vol. 99, pp. 1358-64, 12, 2008.

[7] M. Lokander, and B. Stenberg, "Performance of isotropic magnetorheological rubber materials," Polym. Test., vol. 22, pp. 245-51, 2003.

[8] Z. Varga, G. Filipcsei, and M. Zrinyi, "Magnetic field sensitive functional elastomers with tuneable elastic modulus," Polymer, vol. 47, pp. 227-233, 2006.

[9] T. E. Gómez Alvarez-Arenas, "Simultaneous determination of the ultrasound velocity and the thickness of solid plates from the analysis of thickness resonances using air-coupled ultrasound," Ultrasonics, vol. 50(2), pp. 104–109, 2010.

[10] T. E. Gómez Alvarez-Arenas, "Acoustic impedance matching of piezoelectric transducers to the air," IEEE transactions on ultrasonics, ferroelectrics, and frequency control, 51(5), pp. 624–33, 2004.

[11] T. E. Gómez Álvarez-Arenas, T. R. Shrout, S. J. Zhang and H. J. Lee, "Air-Coupled Transducers Based on 1-3 Connectivity Single Crystal Piezocomposites," 2012 IEEE International Ultrasonics Symposium Proceedings, pp. 2230-2233, 2012.

[12] Z. Hashin, S. Shtrikman, "A variational approach to elastic behaviour of multiphase materials," J. Mech. Phys. Solids, vol. 11, pp. 127, 1963.

[13] T. E. Gómez Alvarez-Arenas, A. Mulholland, G. Hayward, J. Gomatam, "Wave propagation in 0-3/3-3 connectivity composites with complex microstructure," Ultrasonics, 38(9), pp. 897–907. 2000.

T. Nguyen, M. Lethiecq, F. Levassort, L. Pourcelot, "Experimental verification of the theory of elastic properties usisng scattering approximations in (0-3) connectivity composite materials," IEEE Trans. Ultrason. Ferroelec., Freq., Contrl. Vol 43(4), pp. 640-645, 1996.