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A New Constitutive Model for Ferromagnetic Shape Memory Alloy Particulate Composites

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Abstract: Ferromagnetic shape memory alloy particulate composites, which combine the advantages of large magnetic field induced deformation in ferromagnetic shape memory alloys (FSMAs) with high ductility in matrix, can be used for sensor and actuator applications. In this paper, a new constitutive model was proposed to predict the magneto-mechanical behaviors of FSMA particulate composites based on the description for FSMAs, incorporating Eshelby's equivalent inclusion theory. The influencing factors, such as volume fraction of particles and elastic modulus, were analyzed. The magnetic field induced strain and other mechanical properties under different magnetic field intensity were also investigated.

Keywords: ferromagnetic shape memory alloy, particulate composite, Eshelby's equivalent inclusion theory, constitutive model.

1 Introduction

Ferromagnetic shape memory alloys (FSMAs) are a class of intelligent materials with strong magnetic and mechanical coupling, which combine the thermoelastic shape memory effect controlled by temperature with the magnetic shape memory effect controlled by magnetic field. It was reported that the magnet-induced strain of FSMAs achieved can be as large as 10% [O'Handley, Murray, Marioni, Nembach, and Aleen (2000); Sozinov, Likhachev, Lanska, and Ullakko (2002)], and the maximum response frequency can reach 5000Hz [Ezer, Sozinov, and Kimmel (1999)]. In comparison, the response frequency of strain induced by temperature in the traditional shape memory alloys is much lower (less than 1Hz). For FSMA particulate composites, the rigidity of the material and processing performance can be improved by the resin matrix, meanwhile the functionality of the materials can

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be provided by ferromagnetic particles. Thus, FSMA particulate composites possess excellent magneto-mechanical properties and can be widely used in aerospace, automotive and many other fields [Wang and Zhang (2003)].

Shape memory effect can be traced back to the rubber-like behavior, which was found in Au-Cu alloy in 1932. In 1965, Kelly and Davies (1965) firstly proposed the concept of metal matrix composites (MMCs), and then summarized the properties of MMCs. Ferromagnetic shape memory alloys, such as Ni-Mn-Ga [Webster, Ziebeck, and Town (1984)], Ni-Fe-Mn [Liu, Zhang, and Cui (2003)] and Ni-Al-Mn [Morito and Otsuka (1996)], have already aroused much concern. For example, Sato and Okazaki (2002); Zhang, Sato, and Lshida (2006) studied the properties of Co-Ni-Ga alloy. Chernenko, Cesari, and Kokrin (1995) investigated the phase transition temperature of FSMAs. Based on experimental data, Jin, Marioni, and Bono (2002) drew the relation curves of alloying component, martensite transformation temperature and saturation magnetization in Ni-Mn-Ga alloy. Hosoda, Takeuchi, and Inamura (2004) proposed a design procedure for these smart materials and analyzed their shape memory properties. Kainuma, Imano, and Ito (2006) studied the properties of Ni-Co-Mn-Sn Hessler polycrystalline alloys. Dong, Cai, and Gao (2008) investigated the effect of isothermal ageing on the microstructure, martensitic transformation and mechanical properties. Feuchtwanger, Griffin, Huang, Bono, O'Handley, and Allen (2004) studied the mechanical energy absorption in Ni-Mn-Ga polymer composites.

Meanwhile, the constitutive theory for FSMAs was also developed since James's research on magnetostriction of martensite [James and Wutting (1998)]. The constitutive models for FSMAs can be divided into two types: macroscopic models and microscopic models. Macroscopic models can describe the magneto-mechanical properties of FSMAs, while microscopic models can analyze the relations among apparent behaviors and variants, twin crystal or martensite transformation. For example, O'Handley (1988); O'Handley, Murray, Marioni, Nembach, and Aleen (2000) proposed an analytical thermodynamic model for magnetic field induced strain under different anisotropic conditions, outstanding the importance of large magnetic anisotropy and low twin strain to twin boundary motion in FSMAs. Zhu, Chen, and Yu (2014) put forward a three-dimensional quasi-static isothermal incremental constitutive model that was suitable for finite element analysis to study the mechanical behaviors of martensitic variant reorientation. Hirsinger and Lexcellent (2003) developed a non-equilibrium thermodynamic model to express the thermodynamic phase transformation and capture the critical condition for thermodynamic driving of the reorientation in martensite. Based on a tensor description of thermodynamic continuum mechanics taking into account magnetomechanical coupling, Wang and Li (2010) proposed a kinetics model to describe macroscopic behavior of martensitic variants rearrangement in FSMAs.

Although great development has been achieved in FSMAs, less progress can be found in the constitutive modeling of FSMA particulate composites. In this article, a constitutive description for FSMA particulate composites is proposed by combining a simple phenomenological model for FSMA with a mean-field-homogenization approach based on Eshelby's equivalent inclusion theory. The magneto-mechanical behavior of a FSMA particulate composite was also analyzed.

2 Constitutive model

2.1 Description for variants reorientation strain of FSMA

In order to obtain the average strain of particulate composite which is subjected to external magnetic field and pressure, the description for reorientation strain $\boldsymbol{\varepsilon}^r$ of FSMA should be proposed firstly. If FSMA is in martensite state, a single variant (V_M) induced by magnetic field can be obtained when sufficiently large external magnetic field is applied. When compressive stress perpendicular to magnetic field is also applied, twin boundary motion and variants reorientation will occur, and another variant (V_S) can be induced by stress. The volume fraction of V_S will increase with larger stress until the reorientation is finished.

Assuming the maximum reorientation strain is $\boldsymbol{\varepsilon}^r_{\max}$ and FSMA is initially composed entirely of V_S or V_M, the reorientation strain $\boldsymbol{\varepsilon}^r$ can be expressed as [Guo, Li, Wan, Peng, and Wen (2014)]

$$\boldsymbol{\varepsilon}^r = \begin{cases} \xi_H \boldsymbol{\varepsilon}^r_{\max} & \text{initially stress-preferred variant} \\ -\xi_\sigma \boldsymbol{\varepsilon}^r_{\max} & \text{initially field-preferred variant} \end{cases} \tag{1}$$

where ξ_H and ξ_σ are volume fractions of V_M and V_S respectively, and $\xi_H + \xi_\sigma = 1$.

The volume fraction of variants is related to reorientation in martensite. There are some ways to describe the transition. In this work, a hyperbolic tangent function is selected to describe the process of reorientation,

$$\xi_\sigma = \begin{cases} \dfrac{1}{2}\left\{\tanh\left[K_s\left(\sigma_{\mathrm{e}} - \bar{\sigma}_s(\mathbf{H})\right)\right] + 1\right\} & V_M \to V_S \\ \dfrac{1}{2}\left\{\tanh\left[K_m\left(\sigma_{\mathrm{e}} - \bar{\sigma}_m(\mathbf{H})\right)\right] + 1\right\} & V_S \to V_M \end{cases} \tag{2}$$

where $V_M \to V_S$ represents the reorientation from magnetic field induced variant to stress induced variant; σ_{e} is the equivalent stress; K_s and K_m are material constants related to variants reorientation and can be obtained by fitting the stress-strain curve during uniaxial loading; $\bar{\sigma}_s(\mathbf{H})$ is the average value of $\sigma_s^{cr,s}(\mathbf{H})$, the starting critical stress, and $\sigma_s^{cr,f}(\mathbf{H})$, the stress at the end point of the critical range, during the $V_M \rightarrow V_S$ process, that is

$$\bar{\sigma}_s(\mathbf{H}) = \frac{1}{2}\left(\sigma_s^{cr,s}(\mathbf{H}) + \sigma_s^{cr,f}(\mathbf{H})\right) \tag{3}$$

Similarly,

$$\bar{\sigma}_m(\mathbf{H}) = \frac{1}{2}\left(\sigma_m^{cr,s}(\mathbf{H}) + \sigma_m^{cr,f}(\mathbf{H})\right) \tag{4}$$

It is important to note that the average value of critical stress varies with the intensity of magnetic field. Considering the magnetic saturation, the critical stress is nonlinear with the magnetic field and has a saturation value. So the following expression is proposed,

$$\bar{\sigma}_s(\mathbf{H}) = \bar{\sigma}_s^0 + \bar{\sigma}_s^H(\mathbf{H}), \;\; \bar{\sigma}_m(\mathbf{H}) = \bar{\sigma}_m^0 + \bar{\sigma}_m^H(\mathbf{H}) \tag{5}$$

where $\bar{\sigma}_s^0$ and $\bar{\sigma}_m^0$ are mechanical equivalent stresses, $\bar{\sigma}_s^H(\mathbf{H})$ and $\bar{\sigma}_m^H(\mathbf{H})$ are magnetostresses, which are defined as the difference in the stress levels with and without magnetic field.

Usually $\bar{\sigma}_s^H(\mathbf{H}) = \bar{\sigma}_m^H(\mathbf{H})$. Considering the nonlinear property between magnetostress and magnetic field, the magnetostress can be expressed as

$$\bar{\sigma}_s^H(\mathbf{H}) = \bar{\sigma}_m^H(\mathbf{H}) = \bar{\sigma}^H(\mathbf{H}) = \frac{1}{2}\left\{\tanh\left[\mathbf{C}_0(\mathbf{H}-\mathbf{H}_0)\right]+1\right\}\bar{\sigma}_{sat}^H \tag{6}$$

where $\bar{\sigma}_{sat}^H$ is the saturation magnetostress, $\mathbf{H}_0$ is the threshold magnetic field yielding magnetostress, $\mathbf{C}_0$ is the coefficient. Based on equations (1)–(6), the reorientation strain in FSMA can be obtained.

2.2 Constitutive model for FSMA particulate composite

FSMA particulate composite is composed of inclusion phase and matrix phase. To build its constitutive model, the following assumptions are adopted:

(1) The matrix and ferromagnetic particles are isotropic and elastic in the interested range of deformation, and FSMA is in martensite state during the loading process.

(2) The ferromagnetic particles are of sphere with identical size and properties.

(3) The ferromagnetic particles distribute randomly in matrix.

(4) The particles and matrix are bonded perfectly during the deformation.

(5) The magnetic field disturbance and the residual stress induced by the magnetization of particles can be ignored.

If the material is homogeneous and composed of pure matrix, elastic strain $\boldsymbol{\varepsilon}^0$ occurs as external stress is applied. In particulate composite, Mori-Tanaka's theory reveals that disturbance strain $\boldsymbol{\varepsilon}^P$ will be induced when particles are embedded in matrix [Mori and Tanaka (1973)]. In this case, the average stress in matrix can be written as

$$\boldsymbol{\sigma}^0 = \mathbf{L}^0(\boldsymbol{\varepsilon}^0 + \boldsymbol{\varepsilon}^P) \tag{7}$$

where $\mathbf{L}^0$ is the stiffness tensor of matrix.

When external magnet field is applied perpendicularly to pressure, variants reorientation strain $\boldsymbol{\varepsilon}^r$ will appear. According to Eshelby's equivalent inclusion theory [Eshelby (1957)], the average stress in ferromagnetic particles can be expressed as

$$\boldsymbol{\sigma}^1 = \mathbf{L}^1(\boldsymbol{\varepsilon}^0 + \boldsymbol{\varepsilon}^P + \boldsymbol{\varepsilon}' - \boldsymbol{\varepsilon}^r) = \mathbf{L}^0(\boldsymbol{\varepsilon}^0 + \boldsymbol{\varepsilon}^P + \boldsymbol{\varepsilon}' - \boldsymbol{\varepsilon}^*) \tag{8}$$

where $\boldsymbol{\sigma}^1$ and $\mathbf{L}^1$ are average stress and stiffness tensor of particles respectively, $\boldsymbol{\varepsilon}'$ is the difference of mechanical strain in particles and matrix, $\boldsymbol{\varepsilon}^*$ is the equivalent eigenstrain. Eshelby's theory also shows

$$\boldsymbol{\varepsilon}' = \mathbf{S}\varepsilon^* \tag{9}$$

where $\mathbf{S}$ is the Eshelby tensor, which is related to the shape of particle.

The average stress of particulate composite is

$$\bar{\boldsymbol{\sigma}} = \xi_0\boldsymbol{\sigma}^0 + \xi_1\boldsymbol{\sigma}^1 \tag{10}$$

where ξ_0, ξ_1 are volume fractions of matrix and particles respectively, and $\xi_0 + \xi_1 = 1$.

Disturbance strain $\boldsymbol{\varepsilon}^P$ can be derived from equations (7) - (10),

$$\boldsymbol{\varepsilon}^p = -\xi_1(\mathbf{S} - \mathbf{I})\boldsymbol{\varepsilon}^* \tag{11}$$

Equivalent eigenstrain $\boldsymbol{\varepsilon}^*$ can also be obtained from equations (8), (9) and (11),

$$\boldsymbol{\varepsilon}^* = A^{-1}(\mathbf{L}^1 - \mathbf{L}^0)\boldsymbol{\varepsilon}^0 - A^{-1}\mathbf{L}^1\boldsymbol{\varepsilon}^r \tag{12}$$

where $A = [\xi_1(\mathbf{L}^1 - \mathbf{L}^0) + \mathbf{L}^0](\mathbf{S} - \mathbf{I}) - \mathbf{L}^1\mathbf{S}$.

3 Results and discussions

In order to verify the validity of the proposed model, the mechanical behavior of FSMA under external magnetic field is investigated firstly and the prediction is compared with experiment carried out by Couch, Sirohi, and Chopra (2007). The stress-strain curve of FSMA under magnetic field of 6kOe is shown in Fig. 1, where the specimen is subjected to uniaxial loading. The elastic modulus of FSMA is 850 MPa, $\varepsilon^r_{\max} = 5.5\%$, $K_s = 2(\text{MPa})^{-1}$, $K_m = 2(\text{MPa})^{-1}$, $\bar{\sigma}^0_s = 0.4$ MPa, $\bar{\sigma}^0_m = -0.5$ MPa, $C_0 = 0.4(\text{kOe})^{-1}$, $H_0 = 4.2$ kOe, and $\bar{\sigma}^H_{sat} = 3.05$ MPa. It seems that the loading process can be divided into three stages. When compressive stress is applied, the behavior of FSMA is linearly elastic until the stress reaches a critical value, where the twin boundary motion will be induced. The reorientation from variant V_M to variant V_S continues with the increase of compression until second critical stress is achieved. Then FSMA is composed of variants V_S and its behavior is also linearly elastic. The unloading process is similar. It can be seen that response of alloy is pseudoelastic and the prediction of the proposed model shows good agreement with experiment.

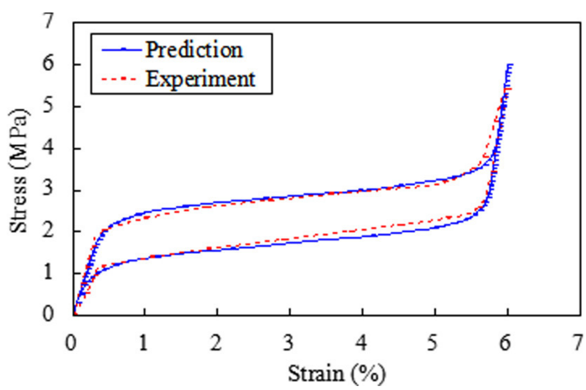

Figure 1: The stress-strain curve of ferromagnetic shape memory alloy under magnetic field of 6kOe.

Assuming the mechanical loading applied on composite is uniaxial and magnetic field is perpendicular to compressive force (see Fig. 2), the magneto-mechanical behaviors of FSMA particulate composite were also analyzed using the proposed constitutive model. The material parameters for FSMA are same as above, meanwhile the elastic modulus and Poisson's ratio for matrix are 6.1MPa and 0.49 respectively.

The FSMA particles are ferromagnetic, and they are embedded in matrix. If FSMA particulate composite is subjected to external magnetic field, the compressive

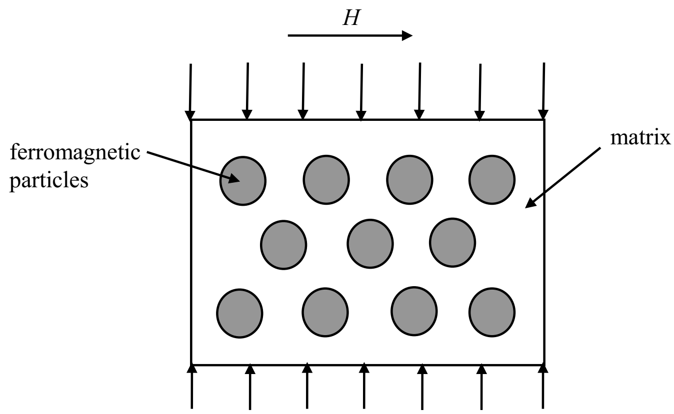

Figure 2: Sketch for the loading on FSMA particulate composite.

stress will appear in particles because of the constraint of matrix. Fig. 3 shows the curve of applied magnetic field versus stress in particles, where no external force is applied on the composites and the volume fractions of particles are different. It is found that the response of particles is nonlinear. The compressive stress will rise with the increase of magnetic field until a saturation value is achieved. It also shows that the saturation value decreases with the increase of volume fractions of particles. It implies that the constraint of matrix becomes weaker when more particles are embedded in matrix. It can be imagined that the stress will be zero if composite is composed of pure FSMA.

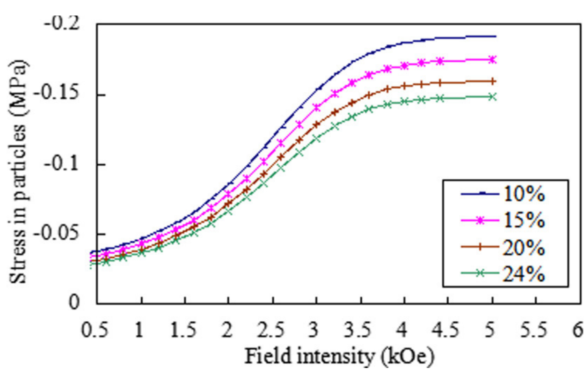

Figure 3: The curve of stress in particles versus magnetic field for composites with different volume fractions of particles.

The magnetic field induced strain in particles is related to the external force and

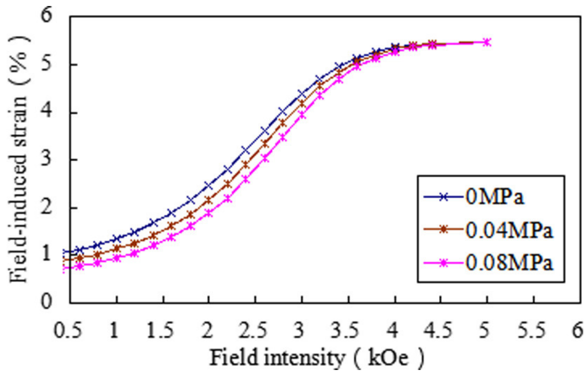

Figure 4: The curve of magnetic field induced strain in particles versus magnetic field under different external pressure.

magnetic field applied on composite. Assuming the composite which composed of 10% volume fraction of particles is subjected to pressure of 0 MPa, 0.04 MPa and 0.08 MPa, respectively, the magnetic field induced strain in particles under different magnetic field is shown in Fig. 4. It can be seen that strain increases with the magnetic field intensity and finally reaches a saturated value. On the other hand, the stronger of external force is applied on composite, the smaller of magnetic field induced strain is produced, except for the case of saturation.

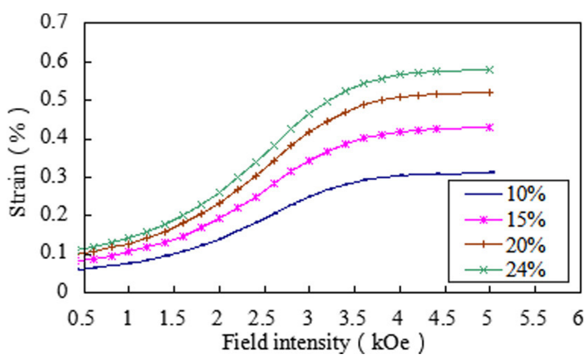

Figure 5: The curve of magnetic field induced strain in composite versus magnetic field with different volume fraction of particles.

Fig. 5 shows the magnetic field induced strain in composites with different volume fraction of particles. Obviously the strain in composites is nonlinear with magnetic field intensity. The magnetic field induced strain increases with field intensity until a saturated value is achieved. It is observed that the strain will be larger for the composite with more FSMA particles. It means that the volume fraction of particles is an important influence factor for the magneto-mechanical performance of composites.

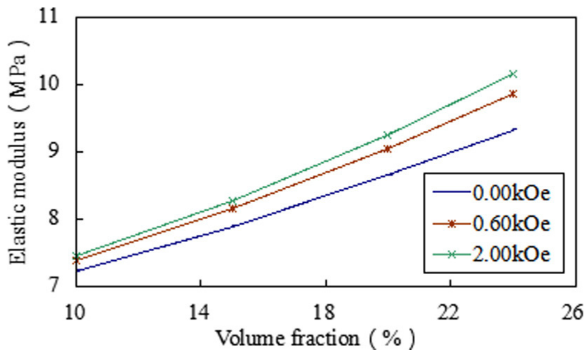

Figure 6: The curve of equivalent elastic modulus of composite versus volume fraction of particles under different magnetic field.

Fig. 6 shows the relation between volume fraction of particles and equivalent elastic modulus of composite under different magnetic field intensity. It is found that the elastic modulus of composite increases with the volume fraction of particles. This agrees with the theory for particulate reinforced composite. Meanwhile the elastic modulus of the composite will increase with the magnetic field intensity due to the magneto-mechanical interaction among FSMA particles.

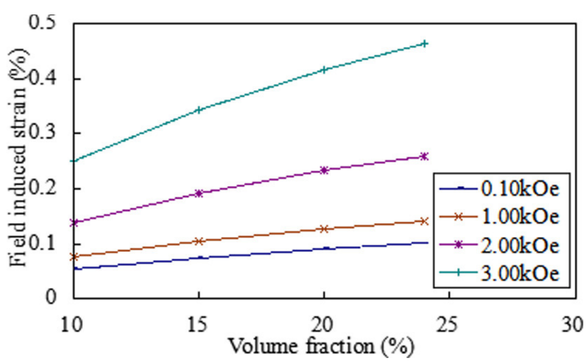

Figure 7: The curve of magnetic field induced strain in composite versus volume fraction of particles.

The volume fraction of particles will affect not only the elastic modulus, but also the magnetic field induced strain in composite, as is shown in Fig. 7. It can be seen that the strain increases with the volume fraction of particles. When stronger magnetic field is applied, the stain will rise more sharply.

4 Conclusions

It is important to describe the behaviors of FSMA particulate composites, especially for the material design and application. In this paper, a new constitutive model, based on the description for variants reorientation in FSMA incorporating Eshelby equivalent inclusion theory, is proposed to analyze the magneto-mechanical performance of the composite. It is found that the stress and strain in particles will increase with the external magnetic field intensity until they reach saturated values. The magnetic field induced strain as well as elastic modulus of composite will also increase if there are more FSMA particles imbedded in matrix.

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References

Couch, R. N.; Sirohi, J.; Chopra, I. (2007): Development of a quasi-static model of NiMnGa Magnetic shape memory alloy. *Journal of Intelligent Material Systems and Structures*, vol. 18, pp. 611–622.

Chernenko, V. A.; Cesari, E.; Kokrin, V. V. (1995): The development of new ferromagnetic shape memory alloys in Ni-Mn-Ga system. *Scripta Metallurgica et Materialia*, vol. 33, pp. 1239–1244.

Dong, G. F.; Cai, W.; Gao, Z. Y. (2008): Effect of isothermal ageing on microstructure, martensitic transformation and mechanical properties of Ni53Mn23.5Ga18.5Ti5 ferromagnetic shape memory alloy. *Scripta Materialia*, vol. 58, pp. 647–650.

Ezer, Y.; Sozinov, A.; Kimmel, G. (1999): Magnetic shape memory (MSM) effect in textured polycrystalline Ni2Mn2Ga. *Proceedings of SPIE*, vol. 3675.

Eshelby, J. D. (1957): The determination of the elastic field of an ellipsoidal inclusion and related problems. *Proceedings of the Royal Society of London. Series A, Mathematical and Physical Sciences*, vol. 241, pp. 376–396.

Feuchtwanger, J.; Griffin, K.; Huang, J. K.; Bono, D.; O'Handley, R. C.; Allen, S. M. (2004): Mechanical energy absorption in Ni–Mn–Ga polymer composites. *Journal of Magnetism and Magnetic Materials*, vol. 272–276, pp. 2038–2039.

Guo, Z. Y.; Li, H. T.; Wan, Q.; Peng, X. H.; Wen, J. (2014): A Model to describe the magnetomechanical behavior of martensite in magnetic shape memory alloy. *Advances in Condensed Matter Physics*, pp. 295436.

Hosoda, H.; Takeuchi, S.; Inamura, T. (2004): Material design and shape memory properties of smart composites composed of polymer and ferromagnetic shape memory alloy particles. *Science and Technology of Advanced Materials*, vol. 5, pp. 503–509.

Hirsinger, L.; Lexcellent, C. (2003): Internal variable model for magneto-mechanical behaviour of ferromagnetic shape memory alloys Ni-Mn-Ga. *Journal De Physique IV,* vol. 112, pp. 977–980.

Hirsinger, L.; Lexcellent, C. (2003): Modelling detwinning of martensite platelets under magnetic and (or) stress actions on NiMnGa alloys. *Journal of Magnetism and Magnetic Materials*, vol. 254–255, pp. 275–277.

Jin, X.; Marioni, M.; Bono, D. (2002): Empirical mapping of Ni-Mn-Ga properties with composition and valence electron concentration. *Applied Physics Letters*, vol. 91, pp. 8222–8224.

James, R. D.; Wutting, M. (1998): Magnetostriction of martensite. *Philosophical Magazine A*. vol. 77, pp. 1273–1299.

Kainuma, R.; Imano, Y.; Ito, W. (2006): Metamagnetic shape memory effect in a Heusler-type Ni43Co7Mn39Sn11 polycrystalline alloy. *Applied Physics Letters*, vol. 88, pp. 192513.

Kelly, A.; Davies, G. J. (1965): The principles of the fiber reinforcement of metals. *International Materials Reviews*, vol. 10, pp. 1–77.

Liu, Z. H.; Zhang, M.; Cui, Y. T. (2003): Martensitic transformation and shape memory effect in ferromagnetic Heusler alloy Ni2FeGa. *Applied Physics Letters,* vol. 82, pp. 424–426.

Morito, S.; Otsuka, K. (1996): Electron microscopy of new martensites with long period stacking order structure in Ni50AlxMn50-x alloys. *Materials Science and Engineering A*, vol. 208, pp. 47–55.

Mori, T.; Tanaka, K. (1973): Average stress in matrix and average elastic energy of materials with misfitting inclusions. *Acta Metallurgica*, vol. 21, pp. 571–574.

O'Handley, R. C. (1988): Model for strain and magnetization in magnetic shape memory alloys. *Applied Physics Letters*, vol. 83, pp. 3263–3270.

O'Handley, R. C.; Murray, S. J.; Marioni, M.; Nembach, H.; Aleen, S. M. (2000): Phenomenology of giant magnetic-field-induced strain in ferromagnetic shape-memory materials. *Journal of Applied Physics*, vol. 87, pp. 4712–4717.

Sato, M.; Okazaki, T. (2002): Development of New magnetostrictive materials in Heusler type CoNiGa system. *Journal of the Japan Institute Metals*, vol. 66, pp. 147–150.

Sozinov, A.; Likhachev, A. A.; Lanska, N.; Ullakko, K. (2002): Giant magnetic-field-induced strain in NiMnGa seven-layered martensitic phase. *Applied Physics Letters,* vol. 80, pp. 1746–1748.

Wang, F. X.; Zhang, Q. X. (2003): Operation principle and application of magnetically controlled shape memory alloy actuators. *Science Technology and Engineer*, vol. 3, pp. 577-581.

Wang, X. Z.; Li, F. (2010): A kinetics model for martensite variants rearrangement in ferromagnetic shape memory alloys. *Journal of Applied Physics*, vol. 108, pp. 113921.

Webster, P. J.; Ziebeck, K. R. A.; Town, S. L. (1984): Magnetic order and phase transformation in Ni2MnGa. *Philosophical Magazine B*, vol. 49, pp. 295–310.

Zhang, J. X.; Sato, M.; Lshida, A. (2006): Deformation mechanism of martensite in Ti-rich Ti-Ni shape memory alloy shin films. *Acta Materialia*, vol. 4, pp. 1185–1198.

Zhu, Y. P.; Chen, T.; Yu, K. (2014): Magneto-Mechanical Finite Element Analysis of Single Crystalline Ni2MnGa Ferromagnetic Shape Memory Alloy. *CMC: Computers, Materials & Continua*, vol. 43, pp. 97–108.