The Effect of Particle Shape on Magnetic Field-Induced Rubber-Like Behavior of Ni-Mn-Ga/Silicone Composites

P. Sratong-on\textsuperscript{1,*}, V.A. Chernenko\textsuperscript{2-5}, and H. Hosoda\textsuperscript{2}

\textsuperscript{1}Automotive Engineering Program, Faculty of Engineering, Thai-Nichi Institute of Technology, 1771/1 Pattanakarn Rd. Suanluang, Bangkok, 10250, Thailand
\textsuperscript{2}Institute of Innovative Research (IIR), Tokyo Institute of Technology, 4259 R2-27 Nagatsuta Midori-ku, Yokohama, 252-8503, Japan
\textsuperscript{3}Ikerbasque, Basque Foundation for Science, Bilbao, 48013, Spain
\textsuperscript{4}BCMaterials, Basque Center for Materials, Applications and Nanostructures, UPV/EHU Science Park, Leioa, 48940, Spain
\textsuperscript{5}University of Basque Country (UPV/EHU), Bilbao, 48080, Spain

* Corresponding Author: pimpet@tni.ac.th

Abstract

The single crystalline of the prototype Ni-Mn-Ga ferromagnetic shape memory alloy exhibits a huge magnetic field-induced strain of 6% due to the re-orientation of twin variants. Therefore, it is promising as the magneto-driven actuator material. However, the magnetic field or mechanical stress applied along the orthogonal direction is necessary to restore the deformed bulk single crystal into the original shape. In order to avoid an application of the extra fields, the Ni-Mn-Ga particles/polymer composite is an alternative solution supported by the concept of the accumulated stress in a polymer matrix which drives a deformation recovery of the composite during switching off of the magnetic field. The objective of this study is to clarify the effect of embedded particle shape on the deformation recovery of Ni-Mn-Ga particles/silicone composites by using the finite element method (FEM) and to investigate the local stress distribution in a polymer matrix between either spherical or rectangular shape particles pairs, whereby to provide the guidelines for design/optimization of the magneto-strain-active Ni-Mn-Ga particles/polymer composites. The case studies of the simulation are divided into an isolated particle and a pair of particles, the particles being positioned either parallel or perpendicular to the applied magnetic field direction. Particularly, the simulations reveal that in case of 200 µm of the inter-particle distance in the pair of spherical particles aligned perpendicularly to the applied field, the polymer layer between particles generates the compressive recovery stress of -0.32 MPa, which is insufficient to restore the deformation of the embedded Ni-Mn-Ga particles during removal of the magnetic field. By contrast, the strain recovery effect can be achieved for the rectangular particle pairs, generating the stress concentration in a matrix of about -0.5 MPa, in a similar condition and arrangement.

Keywords: Ferromagnetic shape memory alloys, Ni-Mn-Ga/polymer composites, Finite element method
1. Introduction
The single crystalline Ni-Mn-Ga martensite, exhibiting a tetragonal structure with lattice parameters ratio of \(c/a < 1\), is a prototype of the ferromagnetic shape memory alloys. It shows 6% of the magnetic field-induced strain due to the twin variant reorientation [1–3]. This deformation can be recovered by either stress or magnetic field applied along the orthogonal direction with regard to the initial direction of the magnetic field. Thus, unlike the conventional shape memory alloys, e.g., Ni-Ti alloys, which exhibit the strain recovery during reverse martensitic transformation [4,5], the actuation of Ni-Mn-Ga is not limited by the heat. As a result, Ni-Mn-Ga is a promising candidate for the magnetically driven high-frequency actuators. However, for such a cyclic actuation, an additional application of the magnetic field or stress along the orthogonal direction is needed to periodically restore the initial shape of Ni-Mn-Ga single crystal. This leads to more complex design and to the efficiency reduction in some applications. Besides, the fabrication of single crystal is a time-consuming and costly process. To avoid these problems, an idea of the fabrication of Ni-Mn-Ga particles from polycrystal [6,7] and their embedding into a polymer, whereby creating a particulate-filled polymer composite, was pursued by many researchers [8–10]. Especially, one of the main advantages of the composite is the storage of stress in the matrix during deformation of an embedded particle under applied magnetic field. The stress in the matrix could be turned into a force to propel shape recovery of individual particles under decreasing of an applied field.

An idea of stress accumulated in the matrix during a mechanical stressing of the Ni-Mn-Ga/silicone composite has been demonstrated in Ref.[11]. Recently, we have shown that elastic force in the matrix which exerted on particles during on/off of the applied magnetic field to the Ni-Mn-Ga/silicone composites depends on the inter-particle distance and particles orientation [11,12]. Therefore, it could be deduced that both the distance- and arrangement-dependent inter-particle interactions cause a non-uniform deformation. This implies the existence of the non-uniform stress fields in the local areas of the matrix, also giving rise to the change in the apparent stiffness of composite, as a global aspect. Hence, we can consider that the non-uniform stress fields in matrix contribute to the strain recovery of Ni-Mn-Ga/polymer composite during removal of the magnetic field. Such a reversible magnetic field induced straining of composite has been found experimentally and called “the magnetic field-induced rubber-like behavior” [12]. It has to be emphasized that this behavior was observed for the irregular shape particles contained in the studied composite [12]. On the other hand, it is known from the bulk of literature on other types of composites that the relationship between particles shape and internal stress considerably affects their mechanical properties and deformation behavior [13–15]. In the case of Ni-Mn-Ga/polymer composite, the influence of the particle shape on the internal stress field in a matrix is unknown and is needed to be clarified.

The objective of this study is to elucidate the effect of particle shape on both the local stress distribution in a polymer matrix and the reversible deformation of Ni-Mn-Ga particles/silicone composites by using a finite element method (FEM). The analysis is performed for the two representative shapes of particles, such as sphere and rectangular prism. The guidelines for a design/optimization of the magneto-strain active Ni-Mn-Ga particles/polymer composites are summarized.

2. Materials and Methods

2.1. Assumptions, modelling and case studies of simulations
The numerical model, used throughout this study, is based on the following assumptions and characteristics of the Ni-Mn-Ga single-crystalline particles [16].

i) The particles have a tetragonal martensitic structure with the lattice parameters ratio \(c/a < 1\) at room temperature. The direction of an easy axis for magnetization is parallel to the \(c\) axis of the crystallographic unit cell.

ii) The principal axes of the cubic unit cell in the parent phase of a particle are parallel to \(x\), \(y\) and \(z\) axes of the orthogonal coordinate system.
iii) Particles contain three different crystallographic directions of martensitic variants. When the magnetic field is applied, one variant, where $c$-axis is parallel to the direction of the applied field grows at the expense of the others.

iv) The deformation of Ni-Mn-Ga single crystal in the martensitic phase follows the plastic deformation criteria. Particularly, the total volume of a single crystal is conserved.

To satisfy the aforementioned assumptions in simulation modelling, the maximum crystallographic strains of Ni-Mn-Ga single crystal represent the strain input parameters along each of the $x$, $y$ and $z$ axes. That is, if the magnetic field is applied along the $x$-axis, -0.06 is the strain input along the $x$-axis, while +0.03 is the strain input along each of the two other orthogonal directions, to conserve the total volume.

In this study, the three representative configurations of the embedded Ni-Mn-Ga particles in the silicone matrix concerning the magnetic field direction are divided into an isolated particle and the pairs of particles that align either parallel or perpendicular directions to the applied magnetic field. The particle pair represents an elementary unit modelling the neighborhood of the close particle in the transversal to the particles chain direction. The spherical and rectangular shapes of particles are considered to investigate the influence of the particle shape on the stress distribution in a matrix. In the simulations, the composite contains a 30vol% of the Ni-Mn-Ga filler with the sizes of spherical and rectangular particles, equal to 100 $\mu$m and 150 $\mu$m, respectively. These particle sizes correspond to the minimum cross-sectional areas described in Refs.[6,12]. All simulation cases in this study are carried out using a commercial finite element software ANSYS 18.0.

The material constants of the Ni-Mn-Ga single crystal are 3100 MPa, 0.33, -0.06 and +0.03 for Young’s modulus in martensitic phase, Poisson’s ratio and crystallographically largest compressive and tensile strain, respectively. The Young’s modulus of 1 MPa is a material constant for the silicone rubber. Since the model of the three configurations of particles is symmetrical, one-eighth of the model with the symmetry boundary conditions are taken into account, as shown in Figure 1(a) for the isolated spherical particle/silicone composite. The one-eighth model with a quadratic hexagonal mesh shape and symmetry boundary conditions in $xy$, $yz$ and $xz$ planes are shown in Figure 1(b). The minimum size of the mesh is 10 $\mu$m.

Figure 1. (a) The case of isolated particle/silicone composite with the one-eighth of model and symmetry boundary conditions. (b) The mesh shape of the one-eighth model of isolated particle/silicone composite. Fine mesh area at the boundary between the Ni-Mn-Ga particle and matrix is illustrated by the green color.
3. Results and Discussions

3.1. Results of simulation

3.1.1. Isolated particle/silicone composite

A 30 vol% isolated particle/silicone composite, containing either spherical or rectangular shape particle (Figure 1), was simulated. Figure 2 illustrates the contour of stress distribution in a matrix of the isolated particle composite along the direction of the applied magnetic field. Isometric view of the model of spherical shape filler is shown in Figure 2(a). Since the matrix is unable to deform under a magnetic field, only particle contracts along the $x$-axis, in direction of the applied field, and elongates in the orthogonal direction. According to Figure 2(a), when particle contracts by 6%, a $-0.45$ MPa of compressive stress, illustrated by red colour, generates at the interface between the spherical particle and matrix. It slightly decreases from the interface to the edge of the composite. In the case of a rectangular particle, the largest compressive stress of $-0.70$ MPa, illustrated by orange colour, generates at the boundary between the rectangular particle and matrix. In addition, it reduces as the distance from the interface increases [16].

The composite can recover its initial shape due to the netback stress, which accumulates in the matrix during application of the magnetic field and releases during removal of the latter. This netback stress is defined by the difference between the absolute value of compressive stress at interface and stress at the edge of the composite. Therefore, netback stresses for one spherical particle (Figure 2a) and one rectangular particle (Figure 2b) in the silicone composite are $-0.33$ MPa and $-0.40$ MPa, respectively. It is known that the stress value close to $-0.5$ MPa, or more, is necessary to trigger a reorientation of the martensitic variants [2,17], hence, higher or equal bias stress for recovery deformation of the single crystal is necessary to be applied in the orthogonal direction. Therefore, the estimated values of the netback stress in the matrix influencing either isolated spherical or isolated rectangular particle is insufficient to recover the shape of the composite.

![Figure 2](image)

**Figure 2.** Stress distribution in the matrix of the 30vol% isolated particle/silicone composite which is parallel to the direction of the applied magnetic field: (a) isometric view of the spherical particle; (b) front view of the isolated rectangular particle. T and C stand for the tensile and compressive stress, respectively.

3.1.2. Configurations of particles pair aligned parallel to the applied magnetic field

Figure 3 illustrates the stress distribution in a matrix containing a pair of particles; both two particles in the pair are either spherical or rectangular, with a 200 μm of the inter-particle distance aligned parallel to the direction of the applied magnetic field. Figure 3(a) shows that, as the particles contract along the applied field direction, this generates $-0.33$–$-0.43$ MPa of the compressive stress (yellow contour) at the interface of particles and matrix which spreads between the two nearby spherical particles. The maximum compressive stress of $-0.64$ MPa (the red color in Figure 3a) occurs at the circumference of a
big circle of spheres. Figure 3(b) shows that -0.35~0.61 MPa of the compressive stress (the orange color) generates at the interface between the rectangular particles and matrix. Unlike spherical particles, compressive stress does not spread between the two rectangular particles, it spreads to the right and left of the particles along the vertical direction (the yellow contour). Since the matrix is incapable to deform by the magnetic field, it tends to resist a contraction of the rectangular particles along the direction of the applied field. As a consequence, a 0.2~0.5 MPa of the tensile stress (the light green color in Figure 3b) occurs in a matrix. Thus, the tensile stress has no significant effect upon the deformation of rectangular particles. By comparison, the spherical particles aligned along the applied field are easier to deform than rectangular particles in a similar configuration, due to the spread of compressive stress field in a matrix along with the particle pairs.

3.1.3 Configuration of particles pair aligned perpendicularly to applied magnetic field

Figure 4 shows the stress distribution in the silicone matrix when the spherical particles are aligned with an orthogonal direction to the applied field. The two inter-particle distances, 200 μm and 400 μm, are considered. Note that the direction of stress distribution shown in Figure 4 is perpendicular to the direction of the applied magnetic field. According to Figure 4, the particles contract along the y-axis and expand along the x-axis. As the horizontal neighbor particles are present, the particles are unable to elongate independently along the x-axis resulting in a -0.33~0.43 MPa of the compressive stress concentration in the matrix between particles illustrated by the yellow and orange colors in Figure 4(a). This compressive stress concentration decreases as the inter-particle distance is greater indicated by the reduction of the yellow color area with a brighter shade as shown in Figure 4(b).

An occurrence of the compressive stress concentration can be explained by the following reasons. During the application of magnetic field in a perpendicular direction to the orientation of pair of particles, the particles contract along the applied field direction (y-axis in Figure 4) and simultaneously elongate along an orthogonal direction to the field (x-axis in Figure 4). In such a configuration, a presence of the horizontal neighbor particles causes difficulties to the expansion of particles. Hence, compressive stress concentration, which opposes the elongation generates in a matrix between the two particles. That is, not only the stiffness of the free matrix constrains the deformation of particles, but also an increase in the stress concentration in the matrix due to the presence of neighbor particles. As a result, the local effective stiffness of composite increases.

Figure 3. Stress distribution in the matrix in case of the configuration when the spherical particles pair (a) or rectangular particles pair (b) are parallel to the applied field. The inter-particle distance is 200 μm. T and C denote a tensile and compressive stress, respectively.
Figure 5 shows the stress concentration in the silicone matrix of either rectangular or spherical particles pairs, oriented perpendicularly to the applied magnetic field, as a function of the reciprocal inter-particle distance. The inter-particle distance is shown along the upper horizontal axis. According to Figure 5, a compressive stress concentration in the matrix in the middle of two nearby particles decreases as the inter-particle distance becomes farther, in a hyperbolic-type manner, for both the rectangular and spherical particles pairs. The compressive stress concentration in a matrix for the rectangular particles pairs is almost two times higher than for the spherical one. The compressive stress concentration decreases to -0.36 MPa at 400 µm of inter-particle distance for the case of the rectangular particles. This value of stress concentration is close to the stress accumulated in a matrix for the isolated rectangular particle/silicone composite. Therefore, the rectangular particles pairs deform independently, similarly to the isolated particle/silicone composite. However, in case of the spherical particles, the maximum stress concentration of -0.32 MPa occurs at 200 µm of the inter-particle distance which is equal to the stress accumulated in a matrix of the isolated spherical particle/silicone composite (Figure 1a). Note that the stress restored in a matrix of the isolated particle/silicone composite is considered to be inadequate to reset the shape of the composite after shut down of the magnetic field. The statement is valid for the case of the spherical particle pairs. In sum, the Ni-Mn-Ga/silicone composites containing rectangular particles aligned perpendicularly to the applied magnetic field with a suitable inter-particle distance are superior to the case of the spherical particles in the same configuration when considering a recovery degree of the deformation of the composite during removal of the magnetic field.

![Figure 4](image_url)  
**Figure 4.** Stress distribution in the silicone matrix in case of pair of spherical particles aligned perpendicularly to the applied magnetic field. The direction of the stress distribution is perpendicular to the applied field. Two distances between the particles, 200 (a) and 400 µm (b), are considered. T and C denote the tensile and compressive stress, respectively.

![Figure 5](image_url)  
**Figure 5.** Compressive stress concentration in the matrix that generates due to the interaction of particles aligned perpendicularly to the applied magnetic field as a function of the reciprocal inter-particle distance. The data for spherical and rectangular particles are shown as the full squares and circles, respectively. The upper horizontal axis shows the inter-particle distance.
4. Conclusions
Recent findings of the giant and recoverable magneto-strain exhibited by the Ni-Mn-Ga particles/silicone composite in Ref. 12 were explained in that work by the outstanding role of the non-uniform internal stress in the matrix accumulated during magnetic field-induced particles straining and released as a driving force to recover the initial shapes of the particles during the switching off of the magnetic field. The stress distribution in the local areas was considered in Ref. 12 as inhomogeneous and highly dependent on the inter-particle interactions and mutual orientation of the particle chains and magnetic field direction. In the present work, by carrying out FEM simulations, we have analyzed the effect of the shape of particles on the internal stress fields generated in the matrix which facilitate the reversible magneto-strain behavior of Ni-Mn-Ga particles/silicone composite. In addition, we have found that the occurrence and change in stress in the matrix depends not only on the shape of neighboring particles, but also their configurations concerning the applied magnetic field. The following results can be highlighted in order to serve as a guide for design of the Ni-Mn-Ga/polymer composite with recoverable deformation under applied/removal magnetic field.

i) Regardless of the shape of the particles, the crystallographic easy magnetization axis (c-axis) in the particles should be aligned perpendicularly to the direction of the applied magnetic field.

ii) When the particles pairs are aligned parallel to the field, they can deform independently resulting in the compressive stress field spread in the matrix (the yellow area in Figure 3(a,b)). Compressive stress field spreads over a wider area of the matrix for the spherical particles than for the rectangular particles. Therefore, a composite made with the spherical particles can transfer the stress via the matrix to particles more efficiently than in the case of rectangular particles.

iii) In the case of complete magneto-strain reversibility, which is called the magnetic field-induced rubber-like behavior of composite [12], the rectangular particles aligned perpendicularly to the applied magnetic field with a suitable inter-particle distance are superior to the case of the spherical particles in the same configuration.

The fabrication method of the magneto-strain active spherical particles of Ni-Mn-Ga is under development. After elaboration of such particles and corresponding measurements, the simulation results will be compared with experiment ones.

Acknowledgments
This research project was supported by Grant-in-Aid of Scientific Research (Kiban S 26220907) from the Japanese Society for the Promotion of Science and by the Spanish Ministry MICINN & U (project RTI2018-094683-B-C53).

References
[1] O’Handley R C Allen S M 2002 Encyclopedia of Smart Materials (New Jersey:John Wiley & Sons, Inc.)
[2] Karaca H E Karaman I Basaran B Chumlyakov Y I Maier H J 2006 Acta Mater. 54 233–245
[3] Pareti L Solzi M Albertini F Paoluzi A 2003 Eur. Phys. J. B - Condens. Matter 32 303–307
[4] Krulevitch P Lee A P Ramsey P B Trevino J C Hamilton J and Northrup M A 1996 J. Microelectromechanical Syst. 5 270–282
[5] Naresh C Bose P S C Rao C S P 2016 Proc. IOP Conf. Ser. Mater. Sci. Eng. 149 012054
[6] Feuchtwaenger J Richard M L Lázpita P Gutiérrez J Barandiarán J M Allen S M O’Handley R C 2008 Mater. Sci. Forum 583 197–212
[7] Nilsén F Aalto I Ge Y Lindroos T Hamnula S P 2015 Mater. Today Proc. 2 S879–882
[8] Hosoda H Takeuchi S Inamura T Wakashima K 2004 Sci. Technol. Adv. Mater. 5 503–509
[9] Glock S Zhang X X Kueza N J Müllner P Michaud V 2014 Compos. Part A Appl. Sci. Manuf. 63 68–75
[10] Glock S Canal L P Grize C M Michaud V 2015 Compos. Sci. Technol. 114 110–118
[11] Sratong-on P Tahara M Inamura T Chernenko V A Hosoda H 2018 Smart Mater. Struct. 27 85024
[12] Sratong-on P Chernenko V A Feuchtwaenger J Hosoda H 2019 Sci. Rep. 2019. 9
[13] Lipetzky P Schmauder S 1994 Int. J. Fract. 65 345–358
[14] Lee B J Mear M E 1999 J. Mech. Phys. Solids. 47 1301–1336
[15] Rasool A Böhm H J 2012 *Int. J. Eng. Sci.* **58** 21–34
[16] Sratong-on P Chernenko V Hosoda H 2019 *Results Mater.* **2** 100037
[17] Straka L Hänninen H Soroka A Sozinov A 2011 *J. Phys. Conf. Ser.* **303** 012079