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Devised New Amorphous Alloys for Magnetoelastic Resonators

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Magnetoelastic Resonator에 사용되는 새로운 비정질 합금

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Abstract

There is a clear and pressing need to reduce bias field (Ha,) used on linear magnetome-chanical resonator tag by at least a factor of two to allow low-bias operation near the frequency minimum since reducing Ha causes a dramatic increase in well depth, which implies increased stability. However, this makes it more difficult to maintain tight frequency specs. It can be solved by a reduction of magnetomechanical coupling (k). We determined from an equivalent circuit model that optimal reduced k is near 0.3. Also, we determined the material properties (λ_s : saturated magnetostriction, M_s, and H_s) that give k=0.3. From these evaluations, we suggested that on optimal composition with adequate material properties is Fe₅₅Co₁₅Cr₆ Nb₂B₁₈Si₄.

1. INTRODUCTION

We have examined the equivalent circuit parameters¹⁻³⁾ to better understand magnetoelastic tag performance and to prepare for an evaluation of the potential of new materials as electronic article surveillance tags^{4,5)}. According to the calculation, deactivatable resonators must show sufficient insensitivity to bias level

that f_r (resonant frequency) falls within a narrow detection range. At the same time the resonant frequency must show sufficient sensitivity to H_b that deactivation (by reducing H_b significantly so f_r rises out of the frequency detection range) is possible. Previously this had been accomplished by operating on an f_r (H_b) curve like that shown as A in Fig. 1. In this case $k \approx 0.5$ –0.6, (k: magnetomechanical

coupling factor), $H_{min} \approx 10-15$ Oe and the operating point, $H_b \approx 6-7$ Oe, left f, too sensitive to bias fluctuations. What is needed is to operate closer to a minimum in $f_r(H_b)$. Simply reducing Ha, gives a curve like B which has a k value of order 0.7; this implies a rapid coupling of mechanical energy to magnetic modes which are highly damped. Thus Q (quality factor) suffers (in a nonresonant system, Q is irrelevant and higher k is always preferred). It is possible to get good amplitude with lower $k \approx 0.3-0.4$, combined with improved stability at fr and adequate deactivation by operating on a curve like C. The reduced k values and maximum magnetization can be found in a range of FeCo-BSi amorphous alloys containing approximately 60 atom % cobalt (e. g. Fe₂₀Co₆₀B₁₈Si₃). However, such cobalt-rich alloys are costly.

Here we describe a new class of amorphous alloys that combines the best physical properties for electronic article surveillance (EAS) sensitivity at reduced coupling values and with minimal use of cobalt. The new alloys are found between the aforementioned FeCoB glasses and the $(Fe_{1-x}[NbCr]_x)_{80}(BSi)_{20}$ glasses, developed for their low ratio of magnetostriction to magnetization. A typical composition in this new class of amorphous alloys is $Fe_{55}Co_{15}Cr_6$ $Nb_2B_{18}Si_4$.

In all cases, it is understood that the metalloid content can vary from a minimum of 14 to 17 atom % (for Fe-rich and Co-rich alloys, respectively) to a maximum of about 26 to 30 atom % (for Fe-rich and Co-rich alloys, respectively). Si should make up not more than one third of the metalloid content in iron-rich alloys to not more than two-thirds in cobalt rich alloys.

2. ISSUES AND CHALLENGES

2. 1 Calculation of Magnetomechanical Coupling Coefficient

Several considerations indicate that electronic article surveillance tags and other magnetoelastic resonating sensors operate best if the magnetomechanical coupling factor is reduced from a value of about k=0.7 (used in many applications) to one of $k\approx 0.3$.

The reason for this recommendation is outlined in Fig. 1. First, it is beneficial to reduce the bias field used in some sensors by at least a factor of two to allow low-bias operation near the more stable frequency minimum (A>B). Simply reducing $H_{\text{\tiny B}}$, causes a dramatic increase in well depth (B). This makes it diffi-

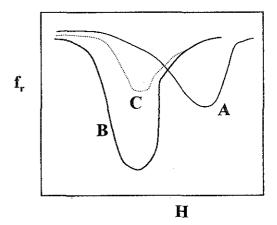


Fig. 1 Schematic of resonance frequency vs. bias field for EAS tags of different anisotropy (A and B) and same anisotropy but different magnetomechanical coupling factor, k (B and C).

cult to maintain tight frequency specs. It has, therefore, been recommended that the magnetomechanical coupling be reduced to an f_r vs. H_b curve as shown in C. It can be determined from an equivalent circuit model⁶⁾ that the desired f_r vs. H_b curve results for $k \approx 0.3$. It is important, therefore, to determine the material properties (λ_b, M_a, H_a) that give k=0.3.

We started by returning to Livingston's simple magnetomechanical equations⁷⁾ for a transverse anisotropy material (i.e. a linear tag). Summarizing results, the magnetomechanical coupling coefficient (measuring fraction of magnetic energy in coupled magnetoelastic modes) is given by

$$k = d(E_H/\chi_o)^{1/2} \tag{1}$$

Here $\chi_a = M_s/H_a$ is the susceptibility at constant stress and, d is the magnetostrictivity which is defined below.

$$d = \partial M/\partial \sigma = \partial e/\partial H = 3\lambda H/H_a^2 \tag{2}$$

EH is the field-dependent Young's modulus:

$$E_{\rm H} = \frac{H_{\rm a}^3 E_{\rm M} M_{\rm s}}{M_{\rm s} H_{\rm a}^3 + 9\lambda E_{\rm M} H^2} \tag{3}$$

 $(E_M$ is the saturated Young's modulus which has a value of about $1.2 \times 10^{12} \, d/cm^2$). We can eliminate d and E_H using Eqs. (1) and (2). Consequently, analysis of magnetomechanical behavior for a transverse anisotropy material indicates that the magnetomechanical coupling coefficient is given by:

$$k^2 = \frac{9\lambda^2 E_M H^2}{M H^3 + 9\lambda^2 E_L H^2} \tag{4}$$

Here, M_s is the magnetization, λ_s is the magnetostriction coefficient, H_a is the anisotropy field, and E_M is the saturated Young's modulus which has a value of about $1.2 \times 10^{12} \, d/cm^2$. This model applies only for fields up to H_a at which point k drops to zero. In the following we assume these values for the parameters: $M_s = 800$ Gauss, $\lambda_s = 12 \times 10^{-6}$, $H_a = 6$ Oe, and $E_M = 1.2 \times 10^{12}$ erg/cm³.

We want to design a resonator that operates at a bias field near the peak in k vs. H (see Fig. 2) and we want this peak value of k to be about 0.3. The peak is rounded on top in a real material. The purpose of the study is to identify a new class of amorphous materials whose parameters can give k=0.3.

From Eq. 4 it is clear that in order to reduce k we should reduce the magnetostriction. We could also reduce k by increasing M_s and this brings with it the added benefit of in-

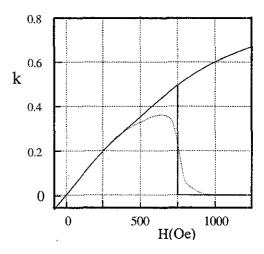


Fig. 2 Variation of magnetomechanical coupling factor, k, with applied field for an anisotropy field of 6 Oe. In a real sample, the peak in k(H) would be rounded as shown by dotted line.

creasing signal strength from the tag. Of course there are physical limitations on the saturation magnetization you can get in an amorphous magnetic material. Here we address the issue of how to combine the greatest possible magnetization and the appropriate magnetostriction to get k=0.3.

The answer to this question lies in solving Eq. 4 for the relation between λ_s and M_s given values for k, H_m , H_s , and E_M :

$$\lambda = \frac{k\sqrt{M_sH_a^3}}{3H\sqrt{E_M(1-k^2)}}\tag{5}$$

$$\lambda_s \approx 2.56 \times 10^{-7} (M_s)^{1/2} \quad (for \quad k = 0.3)$$

Fig. 3 shows plots of λ_s vs. M_s for k=0.3 and 0.4, H=5.5 Oe, $H_s=6$ Oe and $E_M=1.2\times10^{12}$ erg/cm³. To achieve coupling values as low as 0.3, magnetostrictions in the range $6-8\times10^{-6}$

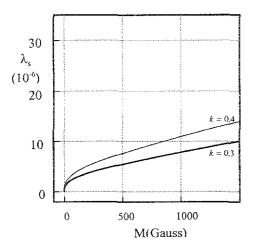


Fig. 3 Lines of constant magnetomechanical coupling in magnetostriction-magnetization space. The lines tell us what combinations of values of magnetostriction and magnetization are required to give k=0.3 and 0.4.

are required if $M_{\rm s}$ is in the range 500 to 1000 G.

2. 2 Control of Anisotropy Field

In any magnetoelastic device it is essential that the anisotropy field, H_a, be controlled because it determines the optimal operating field, slope and magnetomechanical coupling. H_a is governed by alloy composition and by annealing. In general the presence in the material of different magnetic species favors the formation of an induced anisotropy because pair ordering is the microscopic origin of the induced anisotropy and pair ordering at elevated temperatures in a magnetic field is more likely if there are at least two distinct magnetic species. The annealing conditions themselves also determine the strength of the induced anisotropy. Magnetic induced anisotropy vanishes for annealing above the Curie temperature and increases linearly with decreasing temperature just below T_C. However, at sufficiently low temperatures, diffusion cannot occur in reasonable times so the induced anisotropy falls below this linear dependence. It is important to combine optimal composition and processing conditions to obtain the best results.

DISCUSSION

The challenge is to identify the highest-magnetization alloy compositions that fall in the range of values defined in Fig. 3 while making minimal use of cobalt. We want maximum M_s for strong signal output but λ_s low

enough to get $k \approx 0.3$. The answer may be found in some data showing the correlation between λ_s and magnetization in (FeCoNi) $_{80}B_{20}$ metallic glasses8. These data are shown in Fig. 4 as solid dots (FeNi-B20) and open squares (FeCo-B₂₀) superimposed on the model requirement for allowed values of λ_s and M_s for k=0.3 and k=0.4. New alloys meeting the specs should be found in the shaded area which is near Fe₂₀Co₆₀B₂₀. We have indicated the location of some key alloy compositions on these empirical curves. From the point of view of magnetostriction TCA is roughly equivalent to $Fe_{80}B_{20}$, 2826 MB $\approx Fe_{40}Ni_{40}B_{20}$ (A), Co-rich near zero magnetostrictive alloy ≈ Co₇₄Fe₆B₂₀ (B) and CoA ($Fe_{32}Ni_{32}Co_{18}B_{13}Si_{5}$) $\approx Fe_{31}Ni_{31}Co$ $_{18}B_{20}$ (this is located near alloy 6 in Fig. 4). Also added to the same figure are numbered points showing the results of magnetostriction

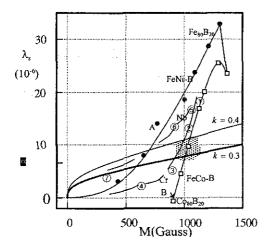


Fig. 4 Same as Fig. 3 but with data superimposed.

Data for amorphous FeNiB alloys (solid dots) and FeCoB (open squares) are shown. Alloys 1-4 are Fess-xCrxBir and 5-7 are Fess-xNbxBir. A=2826MB, B=pinned wall alloy and CoA is located near 6.

and magnetization measurements on the new alloys which are one of the building blocks of the present study. Alloys 1 to 4 are amorphous Fe_{83-X}Cr_xB₁₇ and 5-7 are amorphous Fe_{83-X}Nb_x B₁₇. These Fe (NbCr)B alloys were developed specifically as a low-cost alternative to CoFeB and show reduced magnetostriction with minimal loss of magnetization. Addition of Si for some of the B would improve ribbon quality with little effect on magnetization and magnetostriction.

Clearly the directions we need to go in from 2826MB or CoA alloys are toward higher magnetization and lower magnetostriction. Of the FeCoNi-B choices shown, FeCo alloys offer greater M_s (greater signal) than FeNi alloys. However, to minimize the cobalt content, it is suggested that Nb⁻ and Cr⁻ containing alloys (see numbers 2, 3 and 6 in Fig. 4), plus a minimal amount of cobalt are novel alloys suitable for optimal magnetoelastic response.

A table of possible alloys that combine the best attributes of the Cr and Nb based metallic

Table 1 Recommended alloys of the present invention (atom %).

Fe	Ni	Co	Cr	Nb	В	Si
30	20	28			18	4
30	20	25	3	!	19	4
35	20	20	3		19	3
30	20	22	6		19	3
35	20	17	6		19	3
40	20	12	6		20	2
30	20	25		3	18	4
35	20	20		3	19	3
30	20	22		6	19	3
35	20	17		6	19	3
40	20	12		6	20	2

glasses with those of the cobalt based glasses is included. The first alloy is an average of 2826MB and Co₅₀Fe₄₀B₂₀, but with the metal/metalloid ratio decreased to 78/22. Succeeding alloys seek to minimize the cobalt content by replacement with Cr or Nb. Minor reductions in the Si content are made as cobalt is decreased.

4. CONCLUSION

It is possible to get good amplitude with lower $k \approx 0.3$ –0.4, combined with improved stability at λ_s and adequate deactivation. Based on the equivalent circuit theory, we suggested a new class of amorphous alloys that combines the best physical properties for Electronic Article Surveillance sensitivity at reduced coupling values and with minimal use of cobalt which is costly. The new alloys are found between the conventional FeCoB glasses and the $(Fe_{1-x}[NbCr]_x)_{80}(BSi)_{20}$ glasses, developed for their low ratio of magnetostriction to magnetization. A typical composition in this new class of amorphous alloys is $Fe_{55}Co_{15}Cr_6Nb_2B_{18}Si_4$.

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References

- R. L. Copeland, M. Kopp and R. C. O'Handley, IEEE Trans. Magn., 30 (5) (1994) 3399-3402.
- M. D. Mermelstein, IEEE Trans. Magn. 28
 (1) (1992) 36-56.
- R. C. O'Handley, J. Mater. Eng. Performance,
 (2) (1993) 211-217.
- P. M. Anderson, US Patents, Nos. 4510489: Surveillance System Having Magnetomechanical Marker, 1985.
- P. M. Anderson, US Patents, Nos. 4510490: Coded Surveillance System Having Magnetomechanical Marker, 1985.
- S. Butterworth and F. D. Smith, Phys. Soc., XLIII (2) (1931) 166–185.
- J. D. Livingston, Phys. Stat. Sol. (a) 70, 591 (1982) 591-596.
- 8. O'Handley, Solid St. Comm. 21, (1997) 1119.