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GENERAL ELECTRIC CORPORATE RESEARCH AND DEVELOPMENT --ETC F/G 11/6
INVESTIGATION FOR THE ORIGIN OF MAGNETIC PROPERTIES IN AMORPHOUS--ETC(U)
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**INVESTIGATION FOR THE ORIGIN OF MAGNETIC PROPERTIES
IN AMORPHOUS METALLIC ALLOYS.**

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FOREWORD

This proposal entitled "Investigation for the Origin of Magnetic Properties in Amorphous Metallic Alloys" was prepared as a collaborative project by staff members in the Properties Branch, Metallurgy Laboratory, and the Electronic Power Systems Branch, Electronic Power Conditioning and Control Laboratory, Corporate Research and Development of the General Electric Company.

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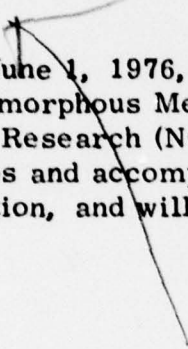
INVESTIGATION FOR THE ORIGIN OF MAGNETIC PROPERTIES
IN AMORPHOUS METALLIC ALLOYS

Section I

INTRODUCTION



The General Electric Research and Development Center started work on amorphous metallic alloys late in 1974. Principal interest has been in the magnetic properties of these alloys. Their mechanical properties, structure, stability and corrosion resistance, and potential applications have also been studied.



On June 1, 1976, a program focussed on "The Origin of Magnetic Properties in Amorphous Metallic Alloys" was begun under sponsorship of the Office of Naval Research (N00014-76-C-0807). This renewal request will review the objectives and accomplishments of the first year's effort and the proposed continuation, and will describe an additional optional effort.

Section 2

DESCRIPTION OF PROJECT

CONTRACT OBJECTIVESGENERAL OBJECTIVE

The general objective of this project is to develop an understanding of the relationships between composition and d-c and a-c magnetic properties, and between composition and stability. It is understood that both composition and preparation parameters may have an influence on the properties, and that both magnetic and structural stability are to be considered.

FIRST YEAR'S EFFORT

1. Stress-relief characteristics and magnetic properties before and after stress relief are being measured for alloys in the following series:

- a. $(\text{Fe}_x\text{Ni}_{1-x})_{80}\text{P}_{14}\text{B}_6$ $\sim 0.3 < x < 1$
- b. $(\text{Fe}_{.5}\text{Ni}_{.5})_{80}\text{P}_{14-x}\text{B}_{6+x}$ $0 < x < 14$
- c. $(\text{Fe}_x\text{Co}_{1-x})_{80}\text{P}_{14}\text{B}_6$ $0 < x < 1$
or $(\text{Fe}_x\text{Co}_{1-x})_{80}\text{B}_{20}$ depending on results obtained in (b)
- d. $(\text{Fe}_x\text{Ni}_y\text{Co}_z)_{80}\text{P}_{14}\text{B}_6$ selected values of x, y, and z
or $(\text{Fe}_x\text{Ni}_y\text{Co}_z)_{80}\text{B}_{20}$ to be determined as above

2. The magnetic properties will include:

Saturation moment, M_s

Coercive force, H_c

Curie temperature, T_c

Remanence to saturation, M_r/M_s

Permeability, μ vs frequency and flux density

Losses, W , vs frequency and flux density

Temperature variation of M_s , H_c , μ , and W

Domain structure

Stress sensitivity of H_c , M_r/M_s , and domain structure

3. The embrittlement behavior will be examined on some of the alloys, to determine the effect of:

- a. Replacing the P by B
- b. Varying the Fe/Ni/Co ratios

ACCOMPLISHMENTS

The first year of this ONR-sponsored work has been extremely productive. Over 50 different compositions from the general system $(\text{Fe, Ni, Co})_x (\text{B, P, Si, C})_y$ have been made into amorphous ribbon. The magnetic properties of these ribbons have been studied in detail, both as made and after various magnetic anneals. Experimental studies have been combined with theoretical analysis to provide an increased understanding of several important areas, including:

- Induced Magnetic Anneal Anisotropy
- Magnetic Stability
- Structural Relaxation and Stability
- Crystallization
- Magnetic Saturation and Curie Temperature
- Losses, Permeability, and Potential Applications

This work has resulted in five papers submitted for publication in the first nine months of the 1976-77 contract period. Preprints of these papers have been sent previously to the ONR Washington and Boston offices. The significant results are summarized in the following paragraphs.

MAGNETIC ANNEAL ANISOTROPY

The uniaxial magnetic anisotropy, K_u , induced by a magnetic anneal has been determined, after stress relief, for a series of alloys given by $(\text{Fe}_y\text{Ni}_{1-y})_{80}\text{B}_{20}$. For all of these alloys K_u depends on anneal temperature as predicted by directional order theory. The concentration dependence of K_u in these alloys is also consistent with directional order theory. The maximum K_u , corrected for its temperature dependence, occurs at the composition $y = 0.5$. However, K_u does not fall to zero at $y = 1$ as predicted if directional order is assumed to arise only from Fe-Ni pair ordering. These results are interpreted as suggesting a role of the glass former, boron, in the directional ordering, perhaps as an interstitial. The interaction energy derived from the results is negative, as expected for interactions leading to precipitation. Its large value of $\approx -7.5 \times 10^{-14}$ ergs corresponds to a critical temperature for precipitation of 3000 K.

MAGNETIC STABILITY

Amorphous $\text{Fe}_{40}\text{Ni}_{40}\text{B}_{20}$ responds to magnetic annealing much more slowly than amorphous $\text{Fe}_{40}\text{Ni}_{40}\text{P}_{14}\text{B}_6$. This implies that the induced anisotropy of the phosphorous-free alloy is considerably more stable. Assuming that the same species are involved in the reordering, this suggests a denser atomic packing in the B alloy than in the P alloy. The amorphous Fe-Ni-B alloy

follows kinetics described either as first order, with a distribution of time constants and activation energies, or as a second order reaction, with a single time constant and activation energy. This is in contrast to the Fe-Ni-P-B alloy, which exhibited first order kinetics with a single time constant and activation energy.

This difference between the phosphorous-free and the P, B-containing alloys is attributed to the atomic uniformity in the reordering environment resulting from the segregation of P. The second order kinetics observed for the reordering of the Fe-Ni-B alloy suggest that two atoms may be involved in the reordering.

STRUCTURAL RELAXATION AND STABILITY

Annealing amorphous ribbons at temperatures below the crystallization temperature induces structural changes in addition to improving the magnetic properties. Specifically, relaxation of internal stresses occurs and the ribbons become brittle. The rate of stress relaxation is the same as the rate of magnetic property improvement; in fact, it is the interaction of the internal strains with the magnetostriction which couples these two effects. The onset of embrittlement with anneal is, on the other hand, a more complex effect. In the phosphorous-containing alloys $(\text{Fe}_x\text{Ni}_y)_{80}\text{P}_{14}\text{B}_6$ embrittlement occurs at very low temperatures of the order of 100°C . In the phosphorous-free alloys $(\text{Fe}_x\text{Ni}_y)_{80}\text{B}_{20}$ embrittlement does not occur until temperatures of the order of 250°C . Prior work in the Research and Development Center indicates that the easy embrittlement of the phosphorous alloys is due to segregation of the phosphorous at low temperatures.

CRYSTALLIZATION

The onset of crystallization has been determined as a function of time and temperature for three amorphous alloys: $\text{Fe}_{40}\text{Ni}_{40}\text{P}_{14}\text{B}_6$, $\text{Fe}_{40}\text{Ni}_{40}\text{B}_{20}$, and $\text{Fe}_{80}\text{B}_{20}$. In addition, the temperature for the onset of crystallization after a two hour anneal has been determined for three series of alloys: $\text{Fe}_y\text{Ni}_{80-y}\text{P}_{14}\text{B}_6$, $\text{Fe}_y\text{Ni}_{80-y}\text{B}_{20}$, and $\text{Fe}_{40}\text{Ni}_{40}\text{P}_{20-z}\text{B}_z$. All of these results have been compared with results reported in the literature for other alloys. An experimental correlation is observed, showing that the activation energy for the initiation of crystallization is proportional to the number of different atomic species in the amorphous alloy. The thermal stability, as measured by ΔE or T_x (2 hours), appears to be proportional to $T_x - T_g$ as expected. The Fe-B amorphous alloy was the least stable of the three alloys studied. It has a projected life of 25 years at 175°C ; adequate for many, but not all, magnetic applications.

MAGNETIC SATURATION AND CURIE TEMPERATURE

The magnetic moment per transition metal atom at 0 K and the Curie temperature were obtained for a series of $(\text{Fe}, \text{Ni})_{80}(\text{P}, \text{B})_{20}$ amorphous

quenched alloy ribbons. Fe/Ni and P/B compositions were varied separately. The moment data can be fitted well by assigning 2.1 Bohr magnetons per Fe atom and 0.6 per Ni atom, with the moment lowered by 0.3 per B atom and 1.0 per P atom. Alternatively, moments varying with composition as shown by neutron diffraction in crystalline alloys, combined with a lowering of 1.2 per B atom and 2.1 per P atom, also fit well. For a given P/B composition, T_c shows a broad maximum at Fe:Ni of about 3:1. For a given transition metal composition, T_c increases with increasing B content.

LOSSES, PERMEABILITY, AND POTENTIAL APPLICATIONS

To date, the amorphous alloys have somewhat higher losses and lower permeabilities than have Fe-Ni alloys of the same thickness, but the amorphous alloys are significantly superior to the Fe-Co and Fe-Si alloys. Applications of the amorphous alloys in small electronic devices appear to be justified where the design optimization can make use of:

- The lower cost expected from the amorphous alloys.
- The higher induction of some of the amorphous alloys compared with the Fe-Ni alloys.
- Their lower losses and higher permeabilities compared with the crystalline Fe-Co and Fe-Si alloys.

The high-saturation amorphous alloys of Fe-B as thin tapes have about one-fourth the losses of the best grain-oriented Fe-3.2% Si sheet steel measured with sine flux, but the saturation magnetization is 20% lower.

The design implications of these differences for power devices is not immediately clear. The temperature dependencies of properties are equivalent to those of the crystalline alloys. The limiting metallurgical life, defined as the start of crystallization, is extrapolated to be 550 years at 175°C and 25 years at 200°C for the least stable, possibly useful alloy tested so far: the Fe₈₀B₂₀.

PUBLICATIONS AND REPORTS RESULTING FROM THE CURRENT INVESTIGATION

1. F.E. Luborsky and J.L. Walter, "Magnetic Anneal Anisotropy in Amorphous Alloys," IEEE Transactions on Magnetics, to appear in March 1977 issue.
2. F.E. Luborsky and J.L. Walter, "Kinetics of Reorientation of the Induced Anisotropy in Amorphous Fe₄₀Ni₄₀B₂₀," Materials Science and Engineering, to appear in 1977.
3. F.E. Luborsky, "Crystallization of Some Fe-Ni Metallic Glasses," Materials Science and Engineering, to appear in 1977.

4. F. E. Luborsky, "Perspective on Application of Amorphous Alloys in Magnetic Devices," to appear in Proceedings of Second International Symposium on Amorphous Magnetism (edited by R. A. Levy and R. Hasagawa), Plenum Press, New York.
5. J. J. Becker, F. E. Luborsky, and J. L. Walter, "Magnetic Moments and Curie Temperatures of $(\text{Fe, Ni})_{80}(\text{P, B})_{20}$ Amorphous Alloys," IEEE Transactions on Magnetics, submitted.
6. F. E. Luborsky, Magnetic Annealing of Metallic Glasses, Gordon Research Conference, July 1976.
7. J. J. Becker, Application of Metallic Glasses as Soft Magnetic Materials, Pittsburgh Section, American Institute of Mining, Metallurgical and Petroleum Engineers, November 5, 1976.

PROPOSED CONTINUATION

The general goals of this work have been to explore, understand, and define the magnetic capability of amorphous alloys. The approach taken is to systematically vary the composition and processing conditions, to measure the magnetic and structural properties both as prepared and after various thermal, mechanical, or magnetic treatments, and to try to optimize the composition and process for potential applications. Significant progress has been made during the first year of this program. Substantial information and some understanding have been achieved in the areas of induced anisotropy, magnetic stability, structural relaxation, crystallization, losses, permeability, saturation, and Curie temperature. This progress, however, has presented several new questions that require further exploration to increase scientific understanding and materials optimization. Most important is an understanding and improvement in the parameters of saturation, losses, permeability, and stability.

The specific approach during the coming year will be to focus on the effect of the metalloids in the $\text{Fe}_x(\text{B, Si, Al, C})_{1-x}$ system. The objectives will be to try to maximize the saturation, stability, and ease of fabrication and minimize the magnetostriction and losses.

SUGGESTED WORK STATEMENT

The following statement is suggested to describe the proposed continuation of work.

1. Magnetic properties and stress relief characteristics will be determined in the iron-rich alloys:
 - a. $\text{Fe}_x\text{B}_{1-x}$
 - b. Fe-B-Si, Fe-B-Si-C, and Fe-B-Si-Al
 - c. Fe-Co-B

2. The magnetic properties will include:

Saturation moment, M_s

Curie temperature, T_c

Coercive force, H_c

Permeability, μ

Losses, W

Magnetostriction, λ

OPTIONAL ADDITIONAL EFFORT: MAGNETIC PROPERTIES
OF CRYSTALLIZED AMORPHOUS MATERIAL

BACKGROUND

During the course of the current studies on crystallization of amorphous metals it was noted that the coercive force increased to 50 to 100 oersteds after crystallization. This observation suggests that in the crystallized form these materials may be ideally useful in hysteresis motor applications.

At present, hysteresis motors use Vanadium Permendur or P-6 alloy for the critical magnetic material. These have a typical composition of 45% Fe, 45% Co, 6% Ni, 4% V and are used in strip form, varying in thickness from 0.006 to 0.100 inch. The cost of these alloys is very high, because of the large amounts of expensive Co, Ni, and V and the additional expense of reducing cast material to thin strip and heat treating. Alternatively, amorphous metals of the $Fe_{80}(B, Si, C, P)_{20}$ family should be considerably less expensive and may perform much better.

The torque developed in hysteresis motors is proportional to the hysteresis loss in the magnetic rotor material. W_h , or the hysteresis loss, is generally considered to be the area of the B-H hysteresis loop. It is desirable to obtain the highest possible value of hysteresis loss. This, of necessity, is limited by the fact that the induction, B, and the field intensity, H, to produce this loss must be supplied by the stator. In choosing the proper H and B and material for a rotor, the designer should consider one more important point: that is, to what degree does the potential magnetic material utilize the peak values of B and H. This can be determined with a comparison of energy factors for the different magnetic materials under consideration.

There are various magnetic materials whose hysteresis loops, when superimposed, could have the same peak values of B and H; however, the hysteresis loss or the areas enclosed by the separate loops for the different materials might vary considerably. For this reason a comparative relationship, known as the energy factor, has been derived to analyze the hysteresis loss of magnetic materials.

The energy factor is determined by constructing a rectangle with sides parallel to the coordinate axes of the hysteresis loop and two opposite corners intersecting the peak points of the hysteresis loop. The ratio of the area of the loop to the area of the rectangle is the energy factor. Materials used in hysteresis motors, for example, generally have energy factors ranging from 0.45 to 0.75. Figure 1 depicts two magnetic materials that have identical peak B and H values but whose loops enclose substantially different areas. Obviously the energy factor of Material No. 1, the solid line loop, is greater than that of Material No. 2. It is therefore desirable to use a material with the highest energy factor possible, to achieve both high motor efficiency and optimum materials utilization.

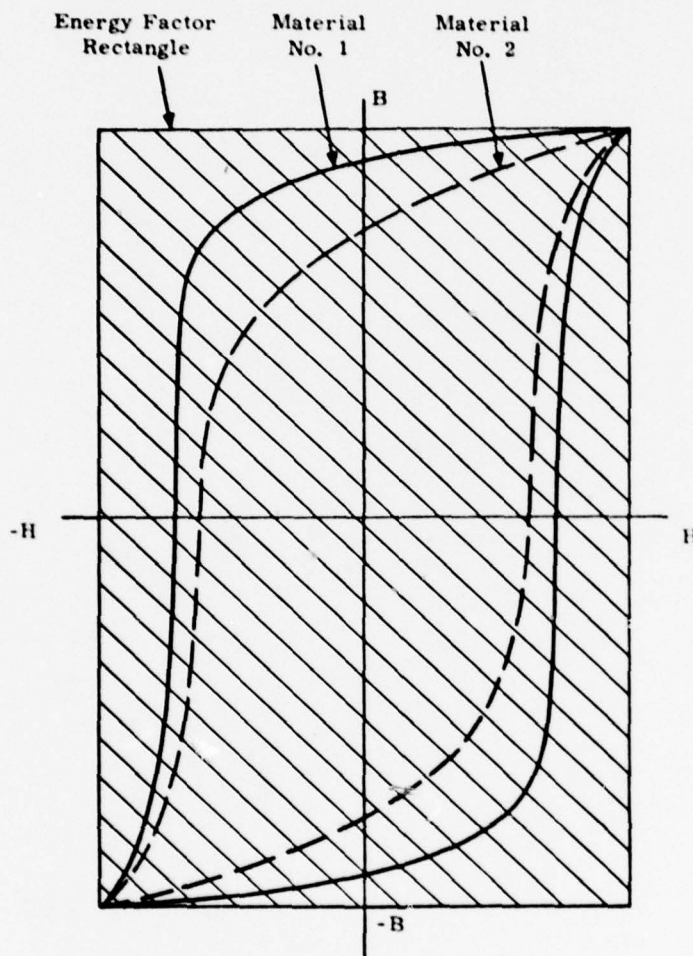


Figure 1. Two Magnetic Materials Having Identical Peak Magnetization Values, but Exhibiting Different Loop Areas

By plotting the energy factor versus peak applied magnetomotive force, the maximum energy factor value is found to occur at the point of maximum

permeability. In most cases, materials with steeper hysteresis loop slopes have been found to have higher energy factors; therefore, for maximum efficiency and minimum size and weight the hysteresis rotor should be used close to the maximum energy factor.

PROPOSED EFFORT

Crystallized amorphous metals of the $Fe_{80}(B, Si, C, P, Al)_{20}$ family appear to have the necessary characteristics for competitive hysteresis material -- low materials cost, low fabrication cost, high residual induction, and proper coercive force. It is proposed that the relevant properties of several compositions from the above alloy family be characterized after a variety of crystallization heat treatments. The hysteresis loops, hysteresis losses, and energy factors will be characterized as a function of applied field, and the composition and heat treatment will be optimized.

PROPOSED WORK STATEMENT

The following work is proposed for the optional additional effort:

1. Magnetic properties of crystallized alloys from the Fe-B-Si-C-Al-P family will be characterized.
2. The composition, process, and heat treatment will be optimized for hysteresis applications.

Section 3

PERSONNEL AND PROGRAM ORGANIZATION

CORPORATE RESEARCH AND DEVELOPMENT ORGANIZATION

General Electric Corporate Research and Development has an objective to provide the technical background and developmental skills which enable Company components to establish new products and businesses. The technical activities of the Center are organized into nine laboratories. Each laboratory is under the direction of a manager responsible for the full spectrum of technical work -- ranging from basic and fundamental research through applied research, feasibility studies, prototype engineering, and the transition stage between development and actual product engineering. The proposed research will be carried out in the Metallurgy Laboratory and the Electronic Power Conditioning and Control Laboratory as a collaborative research project. These two groups have an extensive background in the development of magnetic materials and the conducting of related investigative studies.

PROGRAM ORGANIZATION

The responsibility for the direction and completion of this program will be assumed by Corporate Research and Development. Specifically, Lyman A. Johnson, Manager of the Properties Branch of the Metallurgy Laboratory, will have management responsibility for the work. The principal coinvestigators will be Joseph J. Becker and Fred E. Luborsky. Biographical sketches of the personnel associated with this program follow.

DR. LYMAN A. JOHNSON

Manager - Properties Branch
Metallurgy Laboratory
Corporate Research and Development
General Electric Company

EDUCATION: BA, MA, and PhD in Physical Metallurgy, Harvard University, 1963, 1965, and 1967.

EXPERIENCE: Dr. Johnson has the responsibility for directing the materials-related research and development activities in the technical areas of permanent magnetism, superconductivity, semiconductivity, high-temperature oxidation, and directionally transformed solid-state alloys.

Dr. Johnson joined the research staff of General Electric Corporate Research and Development in 1967. His technical work has included extensive investigations of the mechanisms of metal fatigue and failure and the development and properties evaluation of eutectic alloys for high-temperature structural applications. Dr. Johnson has received two IR-100 Awards for an Electrochemical Crack Detection System, and an Automatic Decompression Computer.

He has been awarded eight U.S. patents. He is a member of the American Society for Metals, and the American Institute of Mining, Metallurgical and Petroleum Engineers, and is a past member of the Publications Committee of Metallurgical Transactions.

DR. JOSEPH J. BECKER

Metallurgist - Properties Branch
Metallurgy Laboratory
Corporate Research and Development
General Electric Company

EDUCATION: BA, MA, and PhD, Harvard University, 1943, 1947, and 1950.

EXPERIENCE: Dr. Becker joined the research staff of General Electric Corporate Research and Development in 1950. His work has dealt with plastic deformation, recrystallization, solid-state precipitation, precipitation hardening, magnetic annealing, permanent magnet materials, losses in soft magnetic materials, kinetics of magnetic domain boundary motion, the morphology of electrochemical electrode processes, and the magnetic properties of amorphous metallic materials.

In the period 1947 to 1950, he held the position of Teaching Fellow at Harvard University.

Dr. Becker has written or coauthored 42 technical papers and articles pertaining to his fields of work. He is a Senior member of the Institute of Electrical and Electronics Engineers and a member of the American Physical Society, the American Society for Metals, and the American Institute of Mining, Metallurgical and Petroleum Engineers. He was chairman of the AIME Electrical and Magnetic Materials Committee, 1957-1960; a member of the AIME Publications Committee, 1961-1964; a member of the Program Committee, Annual Conference on Magnetism and Magnetic Materials in 1958, 1959, 1968, 1970, and 1972; and was AIME Representative for this conference 1957-1960. He was a member of the IEEE INTERMAG Conference Program Committee in 1969. He was Publications Co-chairman for the 1975 Conference on Magnetism and Magnetic Materials, and for the 1976 Joint Conference with INTERMAG, and a member of the Program Committee for both of these conferences.

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DR. FRED E. LUBORSKY

Physical Chemist - Electronic Power Systems Branch
Electronic Power Conditioning and Control Laboratory
Corporate Research and Development
General Electric Company

EDUCATION: BS in Chemistry, University of Pennsylvania, 1947. PhD in Physical Chemistry, Illinois Institute of Technology, 1951.

EXPERIENCE: Dr. Luborsky's main field of interest has been the magnetic properties of materials -- principally hard magnetic materials, thin films, and electrodeposited films. He is currently concerned with high-gradient magnetic separation and studies in amorphous metallic alloys. He has been the key technology leader in developing General Electric's plated-wire memory, magnetic disks, and Lodex[®] permanent magnets.

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Dr. Luborsky has published widely in the fields of plated-wire memories, magnetic thin films, fine particles, and permanent magnet materials and has been awarded ten U. S. patents. Dr. Luborsky is a Fellow of the American Institute of Chemists and the New York Academy of Sciences; a Senior Member of the Institute of Electrical and Electronics Engineers; and a member of the American Chemical Society, the American Physical Society, the American Institute of Physics, the American Association for the Advancement of Science, and the IEEE Magnetics Society.

He has served in many capacities in the INTERMAG and in the Magnetism and Magnetic Materials Conferences as Technical Program Co-chairman, member of the program committee and as local Cochairman. He is serving on the Editorial Board of the International Journal of Magnetism and Magnetic Materials, and is President of the IEEE Magnetics Society.

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