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PRODUCTION AND EVALUATION OF REFE2-NICKEL COMPOSITE MAGNETOSTRI--ETC(U)
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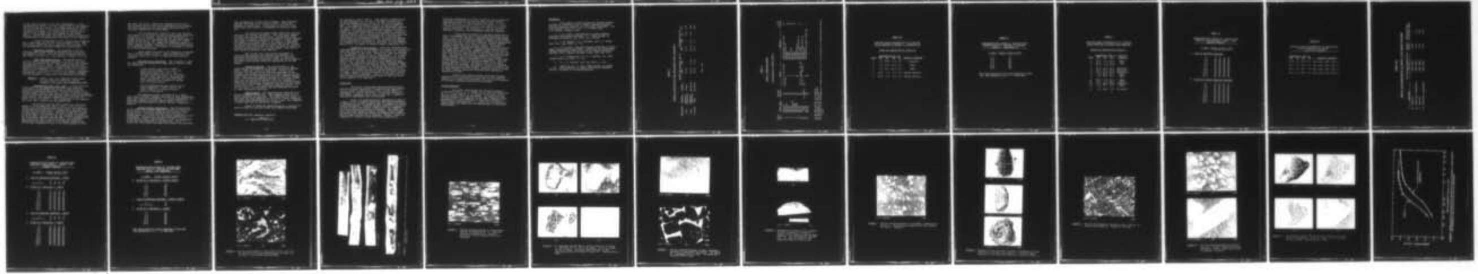

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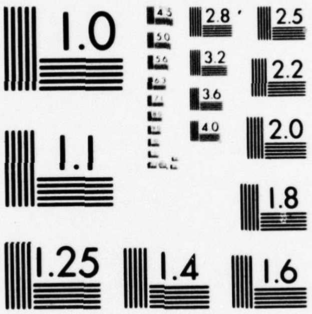

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⑨ FINAL REPORT, TO

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NAVAL ELECTRONIC SYSTEMS COMMAND

CONTRACT NO. N00039-77C0108

⑥ PRODUCTION AND EVALUATION OF $REFe_2$-NICKEL COMPOSITE MAGNETOSTRICTIVE MATERIALS.

Performed by

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PRODUCTION AND EVALUATION OF REFE$_2$-NICKEL COMPOSITE MAGNETOSTRICTIVE MATERIALS

INTRODUCTION

This report describes basic research under Contract No. N00039-77-0108 sponsored by The Naval Electronic Systems Command. The objective was to produce a composite magnetostrictive material consisting of particles of the 'giant' magnetostrictive material, $SmFe_2$, distributed within a pure nickel matrix, and to demonstrate a marked enhancement of the magnetostrictive properties of this composite compared to the properties of nickel. This research is a follow-on to the initial effort in this area performed under Contract No. N00039-76-C-0017.

CONCLUSIONS

1. A composite material containing 20 wt % $Sm_{0.88}Dy_{0.12}Fe_2$ particles distributed in a nickel matrix has exhibited an enhancement of the magnetostriction of the nickel. At an applied field of 12.8 kOe, the composite has 62% increased strain. The strain is still rising rather rapidly at the highest field strength available.

2. The successful composite was made by powder metallurgy techniques, using a combination of room temperature isostatic compaction followed by hot pressing at 1100°F (593°C) or 1200°F (649°C) in a blanked off extrusion press. Hot pressing at 1300°F resulted in excessive interaction of the $SmFe_2$ particles with the nickel matrix.

3. The hot pressed composites are tough and easily machined.

4. Microstructural examination of the successful composite showed the $(Sm,Dy)Fe_2$ particles to be crack-free and to be well bonded to the matrix. Only small quantities of other Sm-Dy-Ni-Fe compounds were found by microprobe analysis. The particles did contain some porosity, apparently due to the presence of divorced eutectic during solidification from the melt.

5. The $Sm_{0.88}Dy_{0.12}Fe_2$ composition is not difficult to melt by induction in a rather large heat. The Dy-containing alloy is more easily homogenized than the $SmFe_2$ composition and requires lower magnetic fields to magnetize.

6. Consolidation of the powder billets by hot extrusion as used in earlier work is not suitable, as previously concluded. The $REFe_2$ type of compound is not sufficiently

plastic at the low extrusion temperatures required to avoid excessive particle/matrix interaction. The higher iron compounds $SmFe_3$ and Sm_2Fe_{17} inadvertently present in initial extrusions do have the necessary plasticity, but are much too difficult to magnetize.

7. Whether or not composites of the type studied here can be considered practical magnetostrictive transducer core materials will depend on a number of factors still to be examined:

- Composites of higher volume fraction of "giant" magnetostriction particles and larger strains than achieved to date will have to be made.
- A practical means of producing laminations of the composite will have to be found to minimize transducer losses.
- Acceptable coupling and efficiency of a working transducer must be achieved.
- The fatigue resistance of the composite must be adequate.
- The cost of the composite material in a suitable product form relative to the derived benefits must be explored for some particular transducer design concept.

BACKGROUND

The initial effort to produce a composite material that would take advantage of the remarkable magnetostrictive strain of the Rare Earth Fe_2 ($REFe_2$) compounds in a ductile and corrosion resistant composite configuration was not successful because of several metallurgical problems (1):

1. Very little of the desired $SmFe_2$ compound was found in the composite. Instead, the higher iron compounds $SmFe_3$ and Sm_2Fe_{17} were identified. These compounds have lower magnetostriction and are more difficult to magnetize than $SmFe_2$. Samarium loss during melting and annealing was the basic problem. Also, because of the small button arc melting procedure used, the particles contained a substantial volume fraction of voids which would be expected to reduce the coupling of the strain to the nickel matrix.

2. Consolidation of powder mixes by high temperature extrusion sometimes resulted in excessive reaction of the Sm-Fe compound particles with the nickel matrix. This was particularly true with the smallest portion of the Sm-Fe

compound particle size distribution. The smallest particles must be several times the reaction zone thickness, found to be about 3μm in the extrusions pressed at 1400°F (760°C).

The earlier work indicated that two other modifications should be made to the procedure:

1. Substitution of a ternary or quaternary RE-Fe compound known to have low anisotropy should allow the use of lower applied fields. The use of $Sm_{0.88}Dy_{0.12}Fe_2$ in a nickel matrix should be attempted; both have negative magnetostriction.

2. Alignment of the $REFe_2$ particles in the powder mix prior to extrusion (or other consolidation technique) might result in the easy magnetization direction <111> being parallel to the working direction axis. This would also lower the magnetic field necessary to magnetize the $REFe_2$ component.

The work reported herein was done in an effort to make composites free of the defects observed in the initial work and thus to test the basic feasibility of a composite of a $REFe_2$ compound in a ductile ferromagnetic matrix. Because the magnetostrictive properties of nickel are well known, nickel was chosen as the matrix. $SmFe_2$, having negative magnetostriction as does nickel, was again chosen as the $REFe_2$ compound. The compound $Sm_{.88}Dy_{.12}Fe_2$, known to have lower magnetocrystalline anisotroy than the binary compound(2), was also used in an attempt to reduce the size of the magnetizing field required to produce useful strain.

In this as well as the previous investigation, a powder metallurgy technique is used to fabricate the composites. A key to success lies in achieving an intimate bonding between the $REFe_2$ particles and the matrix to assure transmission of the huge magnetostrictive strain to the nickel matrix. This bond must be formed by interdiffusion between particle and matrix, but not to such an extent as to change the composition of the bulk of the particle to any marked degree. Net magnetostriction will then depend on the volume fraction of the $REFe_2$ compound incorporated in the composite and the elastic properties of the two phases. Powder metallurgical variables of importance will be: volume fraction $REFe_2$; average particle size and particle size distribution; sintering time and temperature as determined by the consolidation method and conditions chosen; and final density expressed as a percent of the theoretical composite density.

A major problem encountered in attempts to make a suitable composite has been to obtain substantial quantities of the $REFe_2$ alloy compounds of the required composition. Fabrication of these composites requires pounds of material.

Because samarium has a high vapor pressure at the melt and heat treatment temperatures, and because it is extremely reactive with both air and crucible materials, loss of samarium, and thus loss of composition control, has been a major problem. This problem is aggravated by the microstructure resulting from solidification of the alloy and is understandable in terms of the equilibrium diagram (Figure 4 of Ref. 1). The microstructure of the cast alloy will generally contain phases ranging across the entire diagram from nearly pure samarium (solidification of divorced eutectic) to the Sm_2Fe_{17} phase or, in extreme cases, pure iron, at the other end of the diagram. These phases must be eliminated by heat treatment below the peritectic temperature to produce the $SmFe_2$ in the center of the diagram. Considerable long range diffusion is necessary. Loss of samarium to the vapor phase or by oxidation during this lengthy treatment must be avoided.

In the work of this report, we have tried to circumvent the problems discussed in the above paragraph. Arc melting of 50-gram buttons used to produce the Sm-Fe alloy in the first effort was shown in that work to be entirely unsatisfactory(1). In this work, three sources of the alloy are used, and we conclude that melting in rather large quantities is much easier than attempting to make small ingots.

Three series of composites are described in this report.

Series 1. Ni/$SmFe_2$ and Ni/$Sm_{.88}Dy_{.12}Fe_2$ - The Sm-Fe compounds were prepared by Research Chemicals, Phoenix, Arizona. Powder consolidation was by extrusion of canned powder.

Series 2. Ni/$SmFe_2$ - The $SmFe_2$ was prepared at Iowa State University by Dr. Dale McMaster. Powder consolidation was by cold isostatic compaction followed by hot pressing of the powder compact in sealed copper cans.

Series 3. Ni/$Sm_{.88}Dy_{.12}Fe_2$ - The Sm-Dy-Fe compound was induction-melted as a rather large heat at the Inco R&D Center. The same powder consolidation method of Series 2 was used.

Although Series 1 and 2 composites failed to give magnetostrictive strain appreciably greater than that expected of the nickel matrix, the Series 3 composites were found to have nearly twice the saturation strain of nickel. The procedures used in making these composites and the metallurgical and magnetic evaluations of the materials are described below for the three series.

EXPERIMENTAL METHODS

Several aspects of the experimental procedures employed for the three series of composites described in this report are briefly described here; procedures unique to a particular series are described in the text on that series.

Heat Treatment of Cast Sm-Fe Alloys

The inhomogeneous cast alloys must be heat treated at about 850°C, i.e. just below the peritectic at 900°C for the $L + SmFe_3 \rightleftarrows SmFe_2$ transformation. As described above, loss of the volatile and reactive samarium can be a problem. Initially, about 100 grams of coarse chunks of the alloy were placed in glass tubes. Treatments in vacuum, flowing argon, and static argon at about 1/2-atmosphere in a sealed Vycor tube were attempted. All resulted in excessive samarium loss producing alloys containing very substantial amounts of $SmFe_3$, Sm_2Fe_{17}, and even pure iron. The samarium reacts with the glass to form a complex silicate. These problems can be greatly minimized by wrapping the alloy pieces in several layers of tantalum foil, folding over the ends, and sealing the package in Vycor in about 1/2-atmosphere of argon. This was the procedure that was adopted.

The heat treatments were carried out for at least 24 hours and sometimes for several days. The glass capsule was removed from the furnace at the completion of the treatment and allowed to air cool to room temperature.

The alloys were examined by optical microscopy, x-ray diffraction (XRD) and often by electron probe micro-analysis of the phases present before and after heat treatment. Results are presented here where they are instructive.

Preparation of Powder

Heat treated pieces of the cast Sm-Fe alloy were crushed to powder in an argon-filled glove box to prevent the pyrophoric excess samarium (where present) from burning violently during crushing. The powder was screened to isolate the -100 + 325 mesh size fraction in Series 1 and 2 and -100 + 200 in Series 3. This differs from the first attempt where material passing through the 60 mesh screen (250μm), but larger than 325 mesh, was used. The very large particles were found to be unacceptable in the nickel-matrix composite.

Preparation of Billets for Consolidation

The -100 + 325 mesh Sm-Fe powder was blended with

Inco Type 123 nickel powder by weighing and mixing in the argon-filled glove box followed by wire blending in a sealed glass jar. Two loadings, 20 and 40 weight percent, $SmFe_2$ in the nickel were studied.

In the case of Series 1 composites, the blended powder mixes were sealed into appropriate sized copper cans by welding on copper lids. The powder mix was exposed to air in the weld sealing operation. The cans were not evacuated after sealing.

In Series 2 and 3 composites, the powder blend was isostatically compacted at a pressure of 45,000 psi after sealing the powder into plastic bags closed at each end with 4-inch diameter rubber stoppers and hose clamps. This operation produced a solid billet of about 75 to 80% of theoretical density which was then trimmed to fit into the copper container. The copper containers, 3-1/2-inches in diameter x 2-inches long with a 1/4-inch wall thickness and 1/4-inch thick lids, were sealed by heliarc welding.

Consolidation

Series 1 canned billets were consolidated by extrusion at 1400°F (760°C) as described previously(1). Because of difficulties described below with extruded rods, the isostatically compacted and canned billets (Series 2 and 3) were hot compacted by a hot coining operation in the 750-ton extrusion press blanked off to prevent extrusion through a die. Temperatures of 1100°F (595°C), 1200°F (649°C) and 1300°F (704°C) were tried.

Evaluation of Properties

Magnetic moments were determined on 0.1-gram samples in a null coil pendulum magnetometer(3). The specimen and its surrounding coil were located between the tapered pole faces of a 7-inch diameter electromagnet capable of producing magnetic fields up to about 12 kOe.

Magnetostrictive strain was measured on 1-inch long x 1/4-inch x 1/4-inch pieces of the composite by means of strain gauges. The gauge used was Micro-Measurements Type WK-06-125-BT-350, a Ni-Cr alloy gauge of 0.125-inch active length, 350Ω resistance and gauge factor of 2.04. Gauges were bonded to the specimens with M-Bond 200 cement. The flat-sided specimens used here simplified the application of the gauge. The selection of this gauge was based on experience of the earlier measurement where the EA-type self-temperature compensated Constantan (Ni-Cu alloy) gauges were found to exhibit a magnetoresistive effect(1,3,4). The magnetoresistance of the gauge manifested itself as an apparent magnetostrictive strain of the same sign (negative) and of about the same magnitude as the real strain of the nickel or Ni/Sm-Fe specimens. A detailed description of our experience with this class of gauges in large magnetic fields was published in Reference 4. We have suggested that this effect is due to the solid state precipitation of an iron-rich phase and is very sensitive to the heat treatment used to establish the temperature compensation. The WK-type gauges used in the present work are not subject to magnetoresistive effects, but are also more temperature sensitive. All measurements were made at room temperature.

The specimen was mounted in a plexiglass holder by a pin through a hole through the specimen midpoint in a short transverse direction. The holder was then mounted between the pole faces of a 7-inch diameter electromagnet. Magnetic fields to 15.7 kOe were applied parallel to the longitudinal axis of the short bar. The pin mounting prevented the specimen from touching either pole of the electromagnet. Strains were read directly on a Baldwin-Lima-Hamilton SR-4 carrier indicator with both increasing and decreasing magnetic fields in both directions, and in some cases by 90° rotation of the specimens in the field.

Comments on the significance of the 90° specimen rotation measurement are appropriate here.

The change in length, λ, measured in an experiment, depends on the initial and final domain distribution in the specimen according to

$$\lambda = \frac{3}{2} \lambda_s (\langle \cos^2\Theta \rangle_f - \langle \cos^2\Theta \rangle_o)$$

where Θ is the angle between the direction of magnetization and the direction in which the change in length is measured. $\langle \cos^2\Theta \rangle_o$ refers to the initial average domain distribution and $\langle \cos^2\Theta \rangle_f$ to the final. λ_s, the saturation magnetostriction, is a constant of the material. In a relatively soft magnetic material, like nickel in a high field, on rotating the specimen 90°, $\langle \cos^2\Theta \rangle_o$ is unity, $\langle \cos^2\Theta \rangle_f$ is zero, and $\lambda = \frac{3}{2}\lambda_s$. In the present composites, we can assume that the nickel matrix behaves in this way in the high fields necessary to magnetize the $REFe_2$ particles but, because of the very large magnetocrystalline anisotropy of the $REFe_2$ particles, we do not really know for a given field less than the tremendous fields necessary for saturation what $\langle \cos^2\Theta \rangle_o$ and $\langle \cos^2\Theta \rangle_f$ are in the $REFe_2$. We know that they approach unity and zero respectively as the field is increased. In any event, the 90° rotation experiment as used in this work makes it possible to see the contribution to the total change in length of the specimen by the $REFe_2$ portion of the composite above the $\lambda = \frac{3}{2}(37)$ ppm expected of the nickel matrix.

Metallographic examination of the composites was performed using optical microscopy, scanning electron microscopy with energy dispersive chemical analysis, and by wavelength dispersive electron probe microanalysis. This latter technique is particularly valuable in studying the composition of the particles in the composites to determine whether they contain more than one phase, and the nature of the zone of interdiffusion with the matrix. All metallographic specimens were prepared by diamond polishing in a water-free carrier to prevent corrosion of the Sm-Fe phases in water. Examination was usually made in the unetched condition as the Sm-Fe phase is essentially air-etched; the details of the nickel matrix structure are then not visible.

EXPERIMENTAL RESULTS

SERIES 1 - Extrusion of $Ni/SmFe_2$ and $Ni/Sm_{.88}Dy_{.12}Fe_2$

Preparation of Powder Mix. The Sm-Fe alloys were purchased from Research Chemicals of Phoenix, Arizona. The alloys were induction melted in graphite crucibles as heats of about 1200 grams. The yield from these heats was delivered to Inco as broken chunks appropriate for further reduction to powder. The target and actual compositions are listed in Table I. The samarium content is very close to the target in the Sm-Fe alloy, but somewhat low in the Sm,Dy-Fe alloy. Generally, it is wise to melt to a slightly higher samarium level than desired. On equilibration after heat treatment, such an alloy would consist of $SmFe_2$ plus a small amount of samarium from the divorced eutectic rather than $SmFe_2$ plus $SmFe_3$, the situation in samarium-lean equilibrium alloys. The small excess samarium can be expected to be burned in the air when the powder is exposed to air or alloyed with the nickel in the composite; neither situation will harm the composite.

The real problem with the Research Chemical Sm-Fe material (Alloy 1014) is the very segregated microstructure of the as-cast alloy. This structure, shown in Figure 1a, proved to be very difficult to homogenize to the single phase $SmFe_2$ compound by heat treatment. The as-cast structure is dendritic and is clearly multiphase. X-ray diffraction showed that the $SmFe_2$ phase was present with $SmFe_3$, and Sm_2Fe_{17} in substantial amounts. Eutectic samarium is also evident in the microstructure. Heat treatment (Figure 1b) did result in considerable homogenization of the structure, but the Sm_2Fe_{17} and $SmFe_3$ phases are still in evidence by XRD. In addition, Sm_2O_3 was found to be present as well as some iron. The iron formed because of oxidation and volatilization of samarium near the surface of the irregular shaped chunks being heat treated.

The Sm-Fe powder that was finally used to make the composites was largely $SmFe_2$ with some residual evidence of the other compounds. The -100 + 325 screen fraction was blended with nickel powder. The blended powder was packed into copper cans 1-1/2-inches in diameter x 6" long. The 14 extrusions of this series are listed in Table II. Three of the extrusions were made with as-cast Sm-Dy-Fe powder (Alloy

1019) since this powder was not nearly as segregated as the as-cast Sm-Fe alloy 1014 and heat treatment did not materially improve it. Traces of the $SmFe_3$, Sm_2Fe_{17} and Sm_2O_3 were apparent in the x-ray diffraction patterns of both as-received and heat treated material.

Consolidation by Hot Extrusion. The extrusion temperature was 1400°F (760°C) and the extrusion ratio ranged from 5.76:1 to 12.25:1. We had successfully extruded billets with these conditions in the work previously reported(1), but as is apparent in Table II and Figure 2, these extrusions either stalled the press; i.e., were too stiff to extrude through the die, or extruded but emerged with numerous ruptures of the copper canning material and with the Ni/Sm-Fe core severely broken up. We conclude that the core containing $SmFe_2$ is considerably less ductile at a given loading of hard particles and given extrusion temperature than the previous cores that contained (inadvertently) Sm-Fe alloys containing considerably more iron. It may be possible to extrude these powder composites in an extrusion press with more control of ram speed, but with the available unit, the extrusion speed was extremely fast after the first material broke through the die. This explosive action undoubtedly led to tearing of the extruded bar. This series of extrusions was much stiffer than the first. Ductility could be increased by increasing the temperatures, but at the expense of excessive interdiffusion of the matrix and dispersed $SmFe_2$ particles.

Microstructural Examination. Between breaks in the extrusions, there were often sound pieces of consolidated material up to 3 or 4 inches long. A photomicrograph of a longitudinal section of Extrusion #3 is seen in Figure 3. Clearly, microfracturing of the particles is a major problem here. This micrograph is characteristic of all the extruded pieces of this series.

The uncontrolled burst through the extrusion die has another consequence. The high rate of deformation results in near adiabatic heating pushing the temperature of the bar much above the 1400°F (760°C) extrusion temperature. This results in excessive interaction with the matrix and formation of a very thick reaction zone around each particle. This is well illustrated in the specimen current scanning micrograph and scanning x-ray images of Figure 4. The samarium of the original particle is effectively diluted over a much larger volume resulting in substantial departure of the composition from the $SmFe_2$ compound. The electron probe microanalyses shown in Table III show this effect. An extrusive shell of a $SmNi_3$ composition surrounds each particle. Nickel has penetrated to the particle center and samarium has diffused outward leaving the 2-17 phase in the center. This effect was also seen in Billet #1, a composite formed by stalling the extrusion press. This piece did not get as hot as did the extruded rods but considerable reaction with the matrix was evident. Particle fracturing was not a problem in this piece. We conclude that 1400°F (760°C) is too high a temperature to carry out this consolidation process.

Magnetostrictive Strain. The microstructure of Figure 3 would not be expected to show strain enhancement from the Sm-Fe particles in the composite. Clearly the voids in the cracked particles will provide room to absorb the large magnetostrictive strain of the Sm-Fe fragments. The saturation magnetostrictive strains listed in Table II vary from 22 to 45 μin/in. No significant strain enhancement can be claimed. The specimens all reached their saturation strain at the lowest applied field used, 5 kOe, indicating that all the strain is derived from the nickel matrix. Much higher strains would be required to saturate the Sm-Fe particles.

Reasons for the variation in the saturation magnetostriction strain over the range measured have not been examined. One can speculate that variations in compact density may have an effect. Also the reversal of field direction employed in the measurements leads to the distinct possibility that domain rotation in the nickel matrix necessary for magnetostrictive effects may have been minimized by longitudinal domain growth and shrinkage, thus leading to strains lower than the 35 to 40 μin/in expected of pure nickel. A 90° rotation measurement would avoid this problem and was used in one instance with Extrusion #1. The total strain was much the same measured this way (Table IV). Alloying effects in much of the matrix due to the pronounced interdiffusion discussed above would lead to lower average strains.

SERIES 2 - Ni/$SmFe_2$ Composite Consolidated by Cold Isostatic Compaction and Hot Pressing - $SmFe_2$ Prepared at Iowa State University

Preparation of Composites. The difficulties described in the previous section in obtaining a quantity of the $SmFe_2$ compound free of the presence of the higher iron $SmFe_3$ and Sm_2Fe_{17} phases prompted the suggestion by Dr. A. E. Clark of the Naval Surface Weapons Center that we obtain a quantity of the alloy from Dr. Dale McMasters at Iowa State University because of their experience in preparation of these materials. Clark arranged for McMasters to melt 260 g of $SmFe_2$. Four small heats were prepared and sent to IRDC. Three had been annealed for 5 days at 825°C and the fourth for 4 days at 850°C. X-ray diffraction showed only the $SmFe_2$ phase. A small amount of a white phase was seen in the microstructure of the first two heats; the last two, apparently slightly richer in samarium, had eutectic at the grain boundaries. These micrographs are seen in Figure 5. Some porosity is apparent. The quality of the alloys was excellent and provided us with the best material we had to work with up to that time.

The experience in the Series 1 work taught us that the $SmFe_2$ particles do not have sufficient plasticity to withstand powder extrusion at the relatively low temperatures required to minimize gross interdiffusion of the particles and the matrix. Another consolidation technique was required. We decided to try room temperature isostatic compaction followed by hot pressing of the canned compact. The isostatic compaction step produces a solid billet with a density of about 75% of full density and minimizes the volume reduction (and buckling of the protective copper can) experienced in the hot pressing step. The hot pressing step will achieve densification and establish the bond between the matrix and particles with minimum overall change of shape and deformation to fracture the particles.

The $SmFe_2$ was crushed in the usual way and yielded 147 g of -100 + 325 mesh powder. This yielded 735 g of powder when mixed with nickel powder at 20% $SmFe_2$ loading, only enough to make a single billet about 3-inches in diameter and 5/8-inches high after consolidation. The hot pressing temperature was chosen as 1300°F (716°C); 1400°F (760°C) had been shown to be too high in the previous work. The 1300°F temperature turned out to be a poor choice as too much alloying was experienced here also as is shown below. A pure nickel powder compact was prepared in parallel by identical processing.

Pieces cut from the composite hot pressing are shown in Figure 6. The piece had been cut with an ordinary abrasive wheel. The ruggedness and shock resistance of the composite is apparent from the smooth corners and edges.

Microstructural Examination. One sees immediately from the microstructure of the composite, Figure 7, that there has been excessive reaction of the particles with the nickel matrix. Each particle is surrounded by a thick grey reaction zone. Smaller particles are entirely grey. Electron microprobe analysis of a number of particles in the composite are shown in Figure 8 and Table V. A large range of compositions covering possible compounds ranging from the intended $SmFe_2$ to $SmNi_4$ is observed. Very little of the total volume of particles is the $SmFe_2$ composition.

Magnetostrictive Strain. The longitudinal strain measurements on both the composite and the pure nickel reference specimen both made by the isostatic compaction and hot pressing at 1300°F route are presented in Table VI. The field was increased in steps from zero to 13.3 kOe, reduced in steps back to zero, and the cycle repeated with the field in the opposite sense. This is indicated by the arrows over the columns. Note that the pure nickel piece is saturated at the field strength of the first measurement, 1.8 kOe. The saturation strain, -37 ppm is very close to the accepted value for isotropic nickel. The composite does not reach its saturation value on increasing the field until the 3.5 kOe point is reached. The total strain at saturation is again a little lower than that expected of the nickel matrix. Clearly the Sm-Fe particles are making no beneficial contribution to the magnetostictive strain of the composite.

A strain measurement in 90° rotation was attempted, but a lead broke off the last gauge available to us. The Ni-Cr alloy gauges are not as plentiful and available from the manufacturer as the more common Ni-Cu alloy types.

Mechanical Hardness. The hardness of the two specimens used in the strain measurements was measured on the Rockwell B scale. The pure nickel reference specimen was found to be 49.7 R_B. The composite is harder, 99.1 R_B.

High Field Magnetization. The magnetization of the pure nickel reference specimen at 10 kOe at room temperature was found to be 54.8 emu/g indicating this specimen was essentially fully dense. The composite specimen also measured 54.8 emu/g; this is slightly higher than one would expect if the composite is Ni + $SmFe_2$ in that A. E. Clark reports that σ for $SmFe_2$ at 10 kOe is 52.1 emu/g. Apparently the average value of σ at 10 kOe for the several compounds actually present is larger than the magnetization of $SmFe_2$.

SERIES 3 - Ni/$Sm_{0.88}Dy_{0.12}Fe_2$ Composite Consolidated by Cold Isostatic Compaction and Hot Pressing - $Sm_{0.88}Dy_{0.12}Fe_2$ Prepared at INCO

Preparation of $Sm_{0.88}Dy_{0.12}Fe_2$. A 12 kg induction melted heat of the composition was made using samarium purchased from Molybdenum Corporation of America and dysprosium from Research Chemicals. The heat was melted in an alumina-rammed induction box under 1/2-atm. of argon by adding the Sm and Dy after the iron had been melted down and deoxidized under vacuum. The analyzed composition of this heat (T-82138) is given in Table I. This corresponds to the compound $Sm_{0.90}Dy_{.10}Fe_2$, close to the aim composition.

We conclude that the RE-Fe alloys are more easily melted in large quantities than in small. This is particularly true with samarium-containing alloys; the relatively low surface-to-volume ratio of a larger melt minimizes loss of this volatile constituent. Reaction of the melt with the crucible material was minimal, as only 0.015%Al was picked up in the heat. We note too that the (Sm,Dy)Fe_2 compositions are more easily prepared than the $SmFe_2$. This apparently is due to the fact that in the Dy-Fe system the peritectic temperatures are much closer to the liquidus temperatures for a given intermetallic than in the Sm-Fe system; in fact, the $DyFe_3$ and Dy_2Fe_{17} phases are congruent melting(5,6). This prevents much of the severe segregation and necessity for long homogenizing heat treatments characteristic of everyone's experience in making $SmFe_2$.

The microstructure (Figure 9) and x-ray diffraction pattern of the alloy as-cast did show phases other than $(Sm,Dy)Fe_2$ to be present. The alloy was melted with a slight excess of samarium accounting for the black areas of etched-out eutectic. XRD showed the presence of a small amount of the $(Sm,Dy)_2Fe_{17}$ phase and a moderate amount of a phase indexing as $SmFe_5$ according to the calculated pattern of a hexagonal phase(7). This phase is not shown on the Sm-Fe or Dy-Fe phase diagrams.

These phases were easily and completely eliminated with a 26-hour anneal at 850°C. XRD now showed only the $(Sm,Dy)Fe_2$ phase and a trace of samarium from remaining eutectic.

Preparation of Composites. The procedure to make the composites was identical to that used in Series 2 with two exceptions:

- A more limited size range of $REFe_2$ particles was chosen; i.e. -100 + 200 mesh rather than -100 + 325. The smallest particles will be on the order of 74μm rather than 44μm. This step was taken to eliminate the finer particles often observed to be completely reacted with the nickel matrix.
- The hot pressing step was carried out at lower temperature, 1100°F (593°C) and 1200°F (649°C), to minimize matrix-particle interaction.

The composites again contained 20 wt % $REFe_2$. Pure nickel reference compacts were made at each temperature, again with identical processing. In overall appearance, the sectioned composite with the copper canning material in place was indistinguishable from the Series 2 composite of Figure 6.

Microstructural Examination. The microstructure of the 1200°F hot pressed composite is shown in Figure 10. The particles are free of cracks and well bonded to the nickel matrix. The final polishing was done with 1μm diamond paste and accounts for the overall appearance of the micrographs. Although the polished specimens were not intentionally etched, some corrosion in the air contributes to the mottled appearance of the particles. Note, in Figure 10b, that nickel has penetrated between two particles that by chance are rather close together. The pronounced reaction zone seen in the Series 2 1300°F (704°C) hot pressing is absent here. Micrographs of the 1100°F composite were very similar in appearance to those of Figure 10.

The electron microprobe shows (Table VII) that the particles are largely $(Sm,Dy)Fe_2$. Only very small amounts of other iron-rich phases were seen. Occasionally areas very high in samarium, apparently regions of divorce eutectic were noted, as shown in Figure 11 at Point 2. Some nickel is present at all points even in the centers of particles, but the particle/matrix reaction zone, such as the vicinity of Points 1 and 6 of Figure 11, were too narrow to obtain an analysis characteristic of a distinct phase. The width of this region is therefore less than 1μm.

This is the first time we have succeeded in obtaining the composite as originally envisioned; i.e., relatively defect-free particles of $SmFe_2$ or $(Sm,Dy)Fe_2$ uniformly distributed in pore-free nickel and well bonded to the matrix with minimum diffusive interaction of the two phases.

Composite Density. The effectiveness of the consolidation procedure can be assessed by density measurements. These are listed in Table VIII. The 8.9 g/cm^3 obtained for the pure nickel reference specimens is the accepted room temperature density of nickel indicating that these pieces are fully dense with both hot pressing temperatures. The measured composite density of 8.75 g/cm^3 agrees well with the calculated composite density obtained by measuring the density of the $Sm_{0.88}Dy_{0.12}Fe_2$ compound chunks (8.35 g/cm^3) and properly weighting* with that of the nickel matrix (8.9 g/cm^3). The calculated composite density, ρ_C, is 8.78 g/cm^3. On this basis, the hot pressings are 99.7% of their theoretical densities.

Magnetization. The magnetizations measured at 10 kOe are listed in Table VIII. The accepted values for the nickel specimens are obtained. The composites are lower, in fact than the value expected on a Ni/20 wt % $SmFe_2$ composite, indicating that magnetization of the $Sm_{0.88}Dy_{0.12}Fe_2$ compound is lower than that of the Dy-free compound.

Figure 12 shows the magnetization as a function of applied field for the pure nickel and composite specimens

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*Density of a two component composite,

$$\rho_C = \frac{100\rho_1\rho_2}{(\text{wt }\%_1)\rho_2 + (\text{wt }\%_2)\rho_1}$$

hot pressed at 1200°F (649°C). The apparent permeabilities of both specimens are low due to the small specimen and resulting demagnetization effects in this type of measurement. The composite specimen though is less permeable at the lower fields due to the magnetic and mechanical coupling of some of the nickel to the magnetically hard (Sm,Dy)Fe_2 particles. A projected benefit of the composite is that the highly magnetized nickel matrix would provide an effective internal field to aid in magnetizing the (Sm,Dy)Fe_2 particles thus raising the effective permeability of the particle distribution at intermediate fields. We cannot say whether or not this effect is operative. One would have to measure the permeability of the array of particles in a non-magnetic matrix (e.g., Ni-30%Cu) of similar elastic properties, etc.

Magnetostrictive Strain. Strain was measured both longitudinally, i.e., by increasing H in steps in one direction, back down and then up in the other, and also by 90° rotation of the specimen in the field. Results for these two methods are tabulated in Tables IX and X, respectively. It is clear that the nickel reference specimens saturate at the lowest magnetic field applied, $\sim$1.5 kOe, yielding values of 35, 40 and 2/3(58) = 38.7 ppm, just about as we expect for pure nickel. The composites continue to show increasing magnetostrictive effects up to the highest fields available. This is clearly a contribution to the strain due to the (Sm,Dy)Fe_2 portion of the composite; the 35-40 ppm characteristic of the nickel is already attained at applied fields of 2.9 kOe or lower. There is little difference in the 1100°F (593°C) and 1200°F (649°C) specimens. This is consistent with the metallographic observations noted above in that the structures appeared to be identical.

DISCUSSION

The Series 3 experiment described above finally proves the basic feasibility of enhancing the magnetostriction of nickel by distributing within a nickel matrix a dispersion of (RE)Fe_2 particles. To say how effectively the (RE)Fe_2 compounds can be used in this way, a number of additional experiments must be performed.

First, the magnetostriction determined to date is a static value; no dynamic properties have been determined. A composite of suitable geometry must be prepared to perform these measurements. Extended dynamic tests will also serve to tell us about the fatigue properties of the composite particles and particle/matrix interfaces. Will these hold up in long-term dynamic loading? Secondly, higher volume loadings of the (Sm,Dy)Fe_2 should be prepared and tested to see if higher strains can be achieved. Thirdly, since the composite is a metal of high electrical conductivity, practical transducers will have to be built up as stacks of insulated laminations to minimize eddy current losses. Experimental work to date has shown that there is little hope of either hot or cold rolling the composite to sheet. A suitable consolidation technique for making thin sheets must be identified. Hot pressing in a die may suffice, although a vacuum or inert atmosphere would be required.

A significant problem with the $REFe_2$ materials is their low permeability due to high magnetocrystalline anisotropy. Use of the $(Sm,Dy)Fe_2$ compound helps to some extent in principle, but the data presented in Tables IX and X show that the Ni/20 wt % $(Sm,Dy)Fe_2$ composite requires magnetizing fields on the order of at least 10 kOe to derive strains usefully higher than that of pure nickel. This means extremely bulky windings and high currents would be required in an operating transducer. Magnetic field orientation of the $REFe_2$ particles in the composite powder mix to align an easy direction of magnetization in the working direction may help. But besides the intrinsic magnetic properties of the $REFe_2$ compound, the inverse magnetostrictive restraint imposed by the elastic properties of the nickel matrix must be considered a factor in reducing permeability. The high elastic modulus of nickel relative to that of the $REFe_2$ aggrevates this problem. The effect will tend to decrease in severity as the volume fraction of nickel is reduced, but we must conclude that the $Ni/REFe_2$ composites must be viewed as "hard" magnetic materials requiring very substantial magnetizing fields.

Finally the relation between obtainable dynamic magnetostrictive properties and the cost of the composite material in a useful size and shape will have to be examined as the basic composite properties are defined.

ACKNOWLEDGEMENTS

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TABLE I

CHEMICAL ANALYSIS OF THE RARE-EARTH ALLOY MATERIALS

Alloy No.	Source	Alloy	Target RE Content (wt %)		Analysis (wt %)			
			Sm	Dy	Sm	Dy	C	O
1014	Research Chemicals	Sm-Fe	57.39	-	57.4	-	.012	.076
1019	Research Chemicals	Sm,Dy-Fe	50.22	7.40	56.2*	N.D.	.007	.083
T-82138	INCO	Sm,Dy-Fe	50.22	7.40	50.8	7.1	-	.068

*Sm + Dy

TABLE II

SERIES 1 EXTRUSION BILLETS

All preheated to 1400°F (760°C)

Billet No.	Sm-Fe Alloy	Loading (wt %)	Billet	Comments	λ_s x 10^6 μin/in
1	1019 HT(1)	20	1.5" O.D. Cu can through	Stalled press, broke stem	30
2	1019 HT	40	0.43" die	Extruded-broke up	-
3	1019 AC(2)	20	↓	Extruded	42
4	1014 HT	20	↓	Extruded	34
5	1019 HT	20	↓	Stalled press	25
6	1019 HT-MO(3)	20	↓	Extruded	45
7	1019 AC-MO	20	↓	Extruded	45
8	1014 HT-MO	20	↓	Extruded-broke up	-
9	1014 HT	20	↓	Extruded, slower-broke up	-
10	1019 AC	40	1.5" O.D. through .625" die	Stalled press	22
11	1014 HT	40	3" O.D. Cu(4) through 1"	Extruded, slower-broke up	-
12	None	0 (100%Ni)	die	Extruded well	-
13	1014 HT	20	↓	Unconsolidated powder through die	-
14	1019 HT	20	↓	" " " "	-

(1) HT; Heat Treated 24 hrs/850°C.
(2) AC; As-cast - no heat treatment.
(3) MO; Magnetically oriented.
(4) 1.5" O.D. Cu can containing Ni or Ni/Sm-Fe inside 3" O.D. Cu can filled with Cu powder-extruded through 1" die.

TABLE III

ELECTRON PROBE MICROANALYSIS OF A TYPICAL Sm-Fe PARTICLE IN SERIES 1 - EXTRUSION #6

Areas are identified in Figure 4a

	Composition (at. %)				
Area	Fe	Ni	Sm	Dy	Apparent Compound
1	0.2	99.8	0.0	0.0	Ni matrix
2	0.9	76.0	22.8	0.4	$SmNi_3$
3	0.7	76.7	22.4	0.2	$SmNi_3$
4	85.8	3.3	8.4	2.4	$(Sm,Dy)_2(Fe,Ni)_{17}$
5	83.9	5.4	8.5	2.2	$(Sm,Dy)_2(Fe,Ni)_{17}$

TABLE IV

MAGNETOSTRICTIVE STRAIN VS. APPLIED FIELD MEASURED WITH 90° ROTATION OF SPECIMEN

SERIES 1 - EXTRUSION #1

H (kOe)	Strain (in/in x 10^6)*
0	0
1.5	41
2.9	46
5.7	48
8.6	48
11.0	48
12.8	48

*For significance of the strain measured in this way, see explanation on P. 7 of the text.

TABLE V

ELECTRON PROBE MICROANALYSIS OF A TYPICAL Sm-Fe PARTICLE IN SERIES 2 HOT PRESSING

Areas are identified in Figure 8

	Composition (AT. %)			Apparent
Area	Fe	Ni	Sm	Compound
1	64.5	2.1	33.4	$SmFe_2$
2	65.4	3.1	31.6	$SmFe_2$
3	66.7	5.5	27.8	?
4	56.8	18.4	24.8	$Sm(Fe,Ni)_3$
5	62.3	17.4	20.2	$Sm(Fe,Ni)_4$
6	62.6	14.5	22.9	$Sm(Fe,Ni)_4$
7	3.6	63.9	32.5	$SmNi_2$
8	1.6	20.8	77.7	$SmNi_4$
9	62.4	4.4	33.1	$SmFe_2$
10	72.7	3.1	24.2	$SmFe_3$
11	55.6	20.2	24.2	$Sm(Fe,Ni)_3$
12	3.3	74.7	21.9	$SmNi_4$
13	0.9	98.4	0.6	Ni matrix

TABLE VI

MAGNETOSTRICTIVE STRAIN VS. APPLIED FIELD MEASURED LONGITUDINALLY SERIES 2 HOT PRESSED COMPACTS

H (kOe)	Strain (in/in x 10^6)			
	+↓	+↑	−↓	−↑
A. Pure Ni Reference Specimen				
0	0	0	0	0
1.8	36	36	38	38
3.5	36	36	38	38
5.2	36	36	38	38
6.7	36	36	38	38
8.1	36	36	38	38
9.4	36	36	38	38
10.8	36	36	38	38
11.9	36	36	38	38
12.7	36	36	38	38
13.3	36	36	38	38
B. Ni/20 wt % Sm-Fe Composite Specimen				
0	0	0	0	0
1.8	29	32	30	32
3.5	32	32	33	32
5.2	32	32	34	32
6.7	32	32	34	32
8.1	32	32	34	32
9.4	32	32	34	32
10.8	32	32	34	32
11.9	32	32	34	32
12.7	32	32	34	32
13.3	32	32	34	34

TABLE VII

ELECTRON PROBE MICROANALYSES OF FOUR TYPICAL Sm-Dy-Fe PARTICLES IN SERIES 3 1200°F HOT PRESSING

Composition (at. %)				
Fe	Ni	Sm	Dy	Apparent Compound
64.9	1.4	29.6	4.1	$Sm_{0.89}Dy_{0.12}Fe_{1.95}Ni_{0.04}$
65.3	1.4	30.1	3.2	$Sm_{0.90}Dy_{0.10}Fe_{1.96}Ni_{0.04}$
65.3	1.4	29.7	3.6	$Sm_{0.89}Dy_{0.10}Fe_{1.96}Ni_{0.04}$
65.2	1.4	29.2	4.3	$Sm_{0.88}Dy_{0.13}Fe_{1.96}Ni_{0.04}$

TABLE VIII

DENSITY AND MAGNETIZATION OF SERIES 3 HOT PRESSED SPECIMENS

Specimen	Hot Pressing Temperature °F (°C)	Density g/cm^3	Magnetization at 10 kOe, emu/g
A. Pure Ni	1100 (593)	8.90	-
B. Ni/20 wt % $(Sm,Dy)Fe_2$	1100 (593)	8.75	51.1
C. Pure Ni	1200 (649)	8.90	54.1
D. Ni/20 wt % $(Sm,Dy)Fe_2$	1200 (649)	8.75	51.6

TABLE IX

MAGNETOSTRICITVE STRAIN VS. APPLIED FIELD MEASURED LONGITUDINALLY. SERIES 3 HOT PRESSED COMPACTS

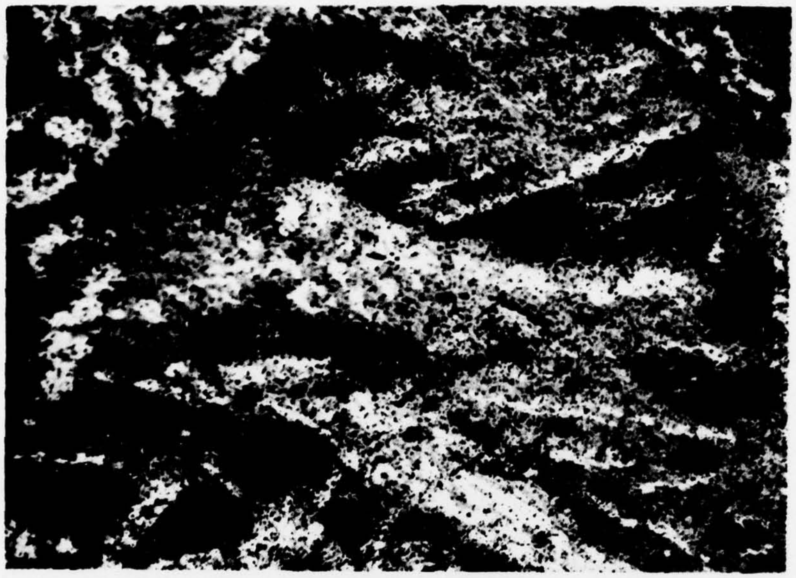
H (kOe)	Strain (in/in x 10^6)			
	+↓	+↑	−↓	−↑
A. Pure Ni Reference Specimen - 1100°F				
0	0	0	0	0
1.8 to 13.3	40	40	40	40
B. Ni/20 wt % (Sm,Dy)Fe_2 - 1100°F				
0	0	0	0	0
1.8	37	37	36	36
3.5	40	40	39	39
5.2	42	43	41	41
6.7	46	47	45	46
8.1	50	49	49	48
9.4	52	51	50	49
10.8	53	53	51	52
11.9	57	57	55	55
12.7	59	59	57	56
13.3	59	59	58	58
C. Pure Ni Reference Specimen - 1200°F				
0	0	0	0	0
1.8 to 13.3	35	35	35	35
D. Ni/20 wt % (Sm,Dy)Fe_2 - 1200°F				
0	0	0	0	0
1.8	40	43	38	40
3.5	46	48	42	43
5.2	49	50	45	47
6.7	52	55	51	51
8.1	56	58	54	55
9.4	60	60	59	57
10.8	64	64	60	60
11.9	66	67	64	63
12.7	68	69	65	64
13.3	70	70	66	66

TABLE X

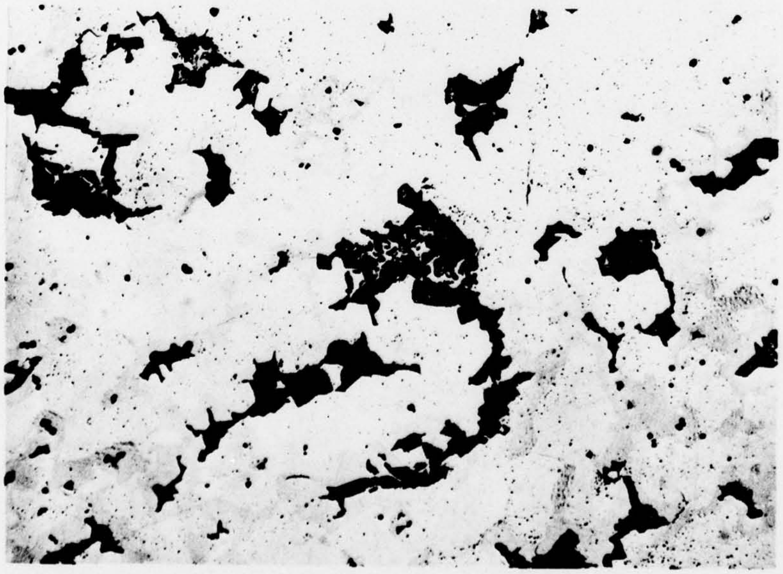
MAGNETOSTRICTIVE STRAIN VS. APPLIED FIELD MEASURED WITH 90° ROTATION OF SPECIMENS SERIES 3 HOT PRESSINGS

	H (kOe)	Strain (in/in x 10^6)*
B.	Ni/20 wt % $(Sm,Dy)Fe_2$ - 1100°F (593°C)	
	0	0
	1.5	48
	2.9	56
	5.7	67
	8.6	78
	11.0	83
	12.8	89
C.	Pure Ni Reference Specimen - 1200°F (649°C)	
	0	0
	1.5 to 12.8	58
D.	Ni/20 wt % $(Sm,Dy)Fe_2$ - 1200°F	
	0	0
	1.5	49
	2.9	58
	5.7	69
	8.6	76
	11.0	82
	12.8	88

*For significance of strain measured in this way, see explanation of P. 7 of the text.

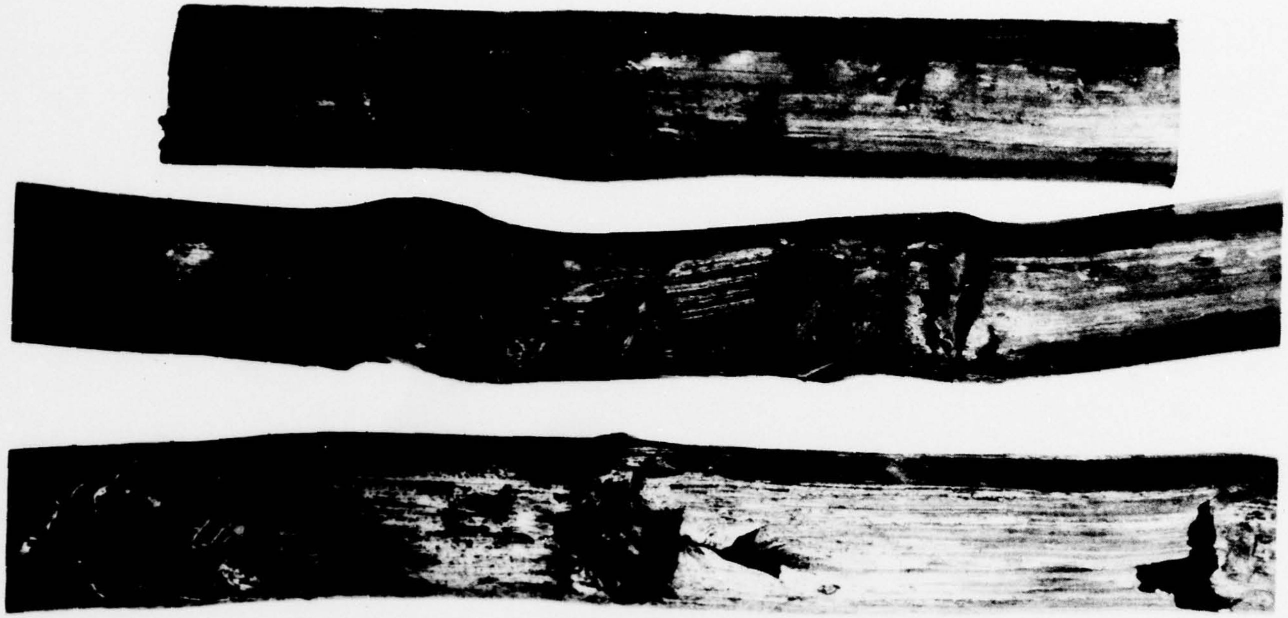


P.N. 1-62791 (a) 200X



P.N. 1-66131 (b) 200X

FIGURE 1 - Optical micrographs of Alloy 1014, Fe-57.4 wt % Sm. (a) as-cast; (b) after homogenization at 850°C for 24 hours. Dry polished and unetched.



P.N. 2-74858

1X

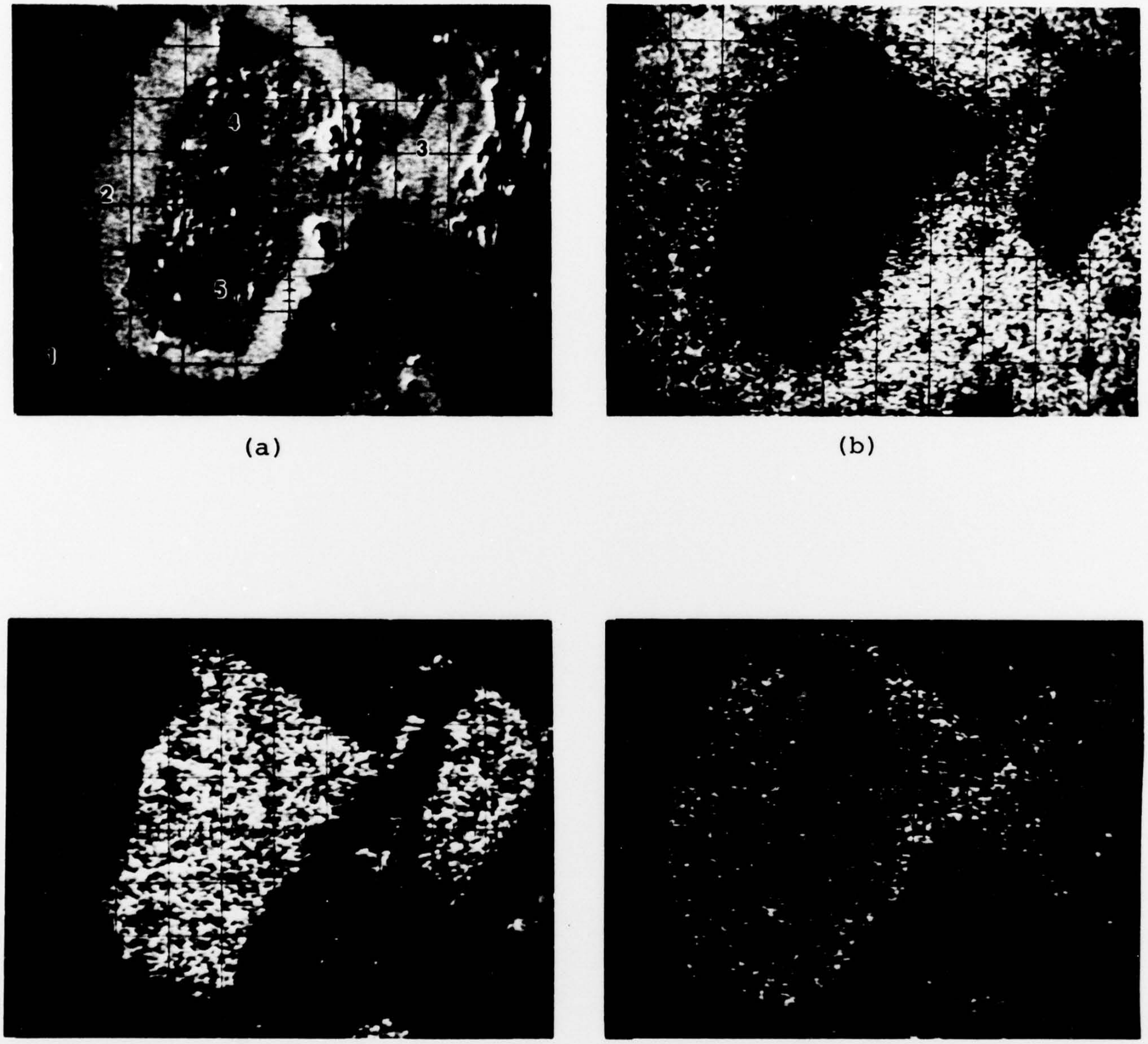
P.N. 2-74855

1X

FIGURE 2 - Macrographs of Series 1 extrusions showing numerous breaks and ruptures through the copper sheath.

P.N. 1-70891 100X

FIGURE 3 - Optical photomicrograph of longitudinal section of Extrusion No. 3. Note extensive microcracking of Sm,Dy-Fe particles. 20 wt % Alloy 1019 in Ni. Unetched.



(a)

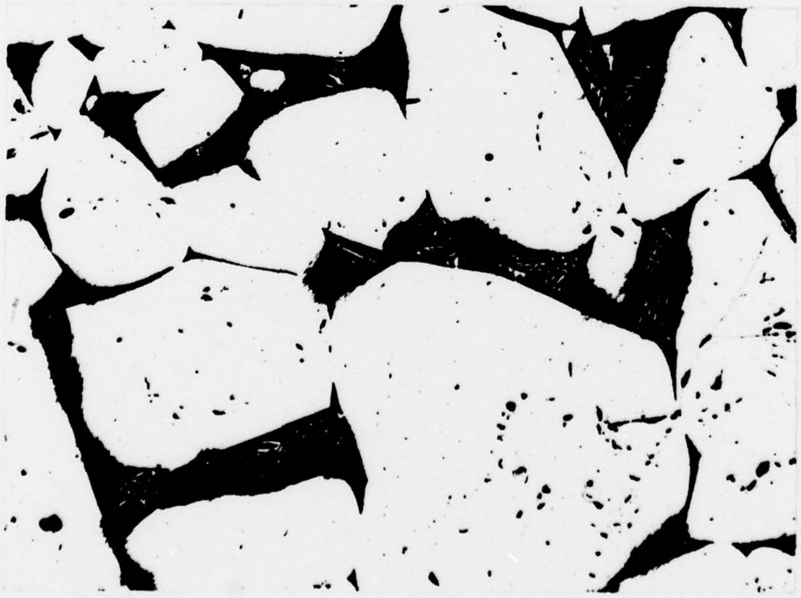
(b)

(c)

(d)

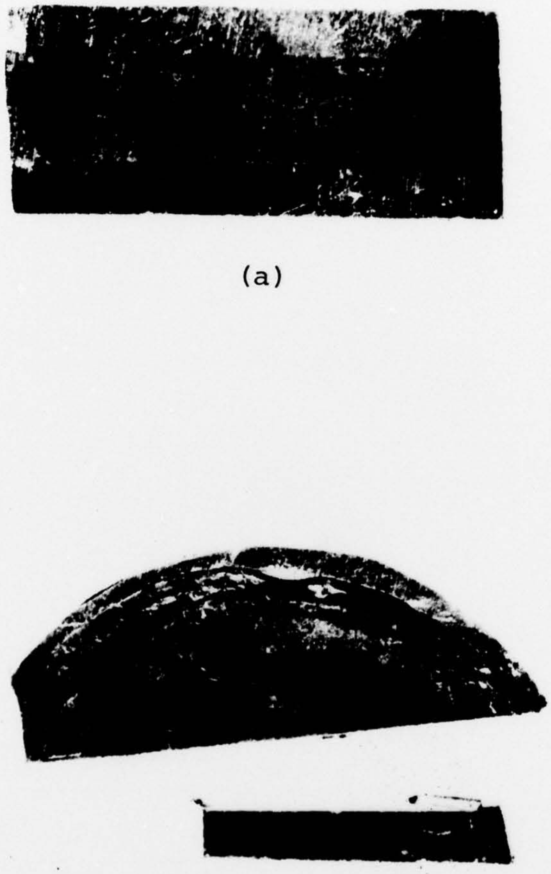
<u>FIGURE 4</u> - (a) Specimen current SEM of typical particle in Series 1 - Extrusion No. 6, (b) Ni K_α image. (c) Fe K_α image. (d) Sm L_α image. Electron probe analyses at the indicated points are listed in Table III. Magnification = 500X.

P.N. 1-73467 (a) 100X



P.N. 1-73470 (b) 100X

FIGURE 5 - Optical photomicrographs of $SmFe_2$ compounds prepared at Iowa State University. (a) Batch No. 1 annealed 825°C/5 days. (b) Batch No. 4 annealed 850°C/4 days.



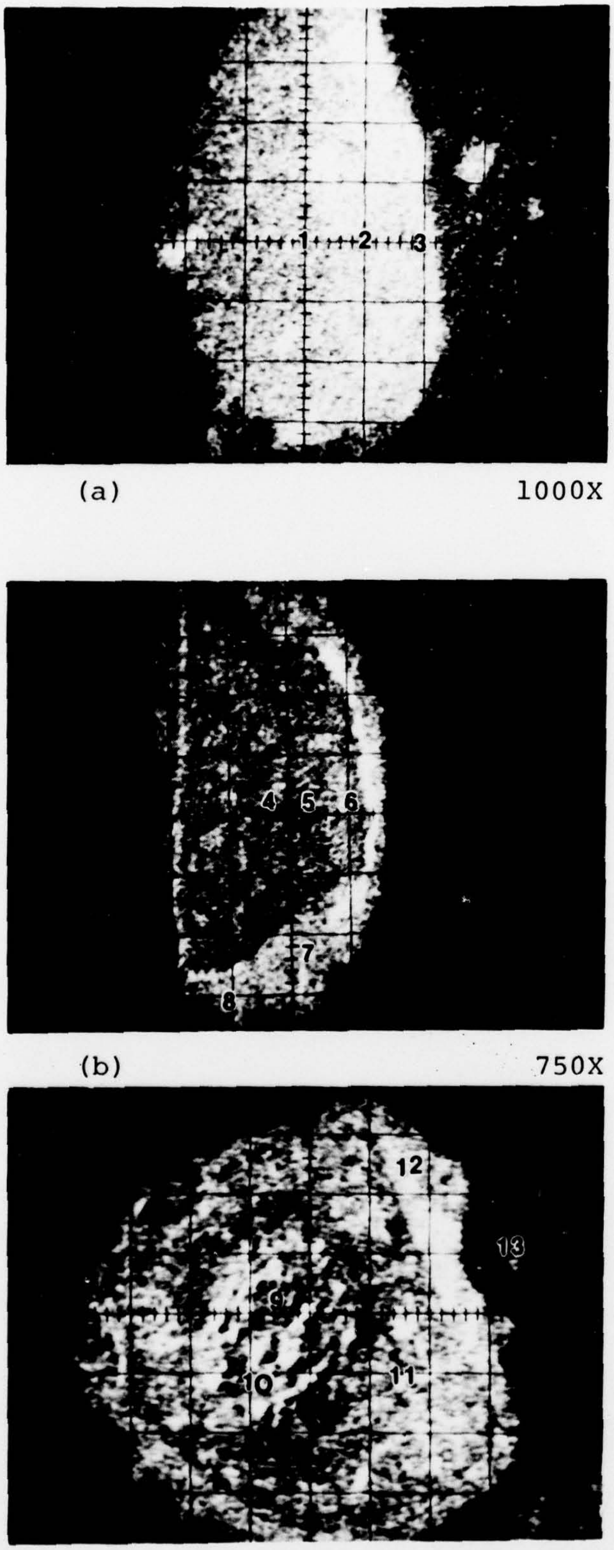
(a)

P.N. 2-74857 1X

FIGURE 6 - Macrograph of pieces of hot pressed Ni/$SmFe_2$ composite of Series 2. Copper canning material has not been removed. (a) Cross-section through can. (b) View of pieces of hot pressing normal to view in (a).

P.N. 1-78148 100X

FIGURE 7 - Optical photomicrograph of Ni/20%$SmFe_2$ composite of Series 2. Isostatically compacted and hot pressed at 1300°F. Unetched.

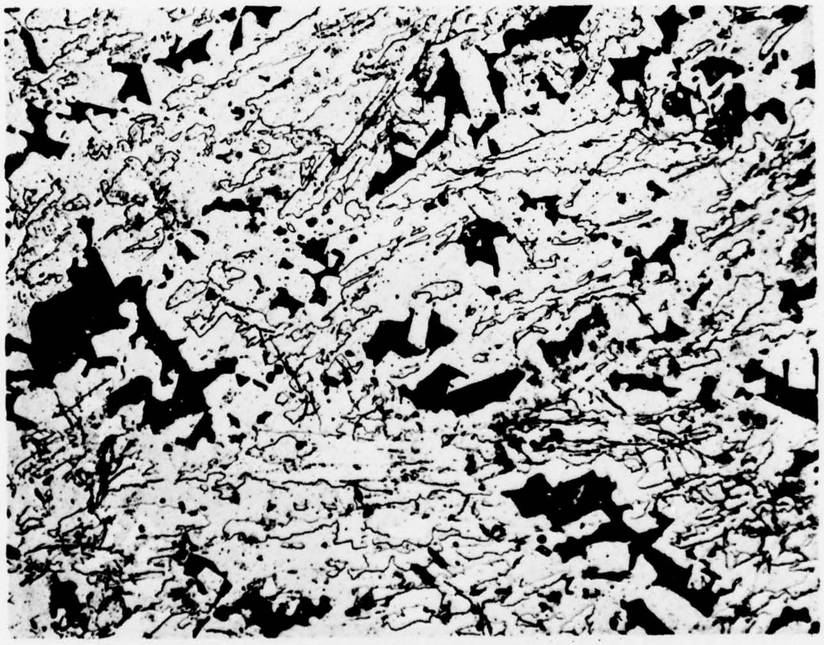


(a) 1000X

(b) 750X

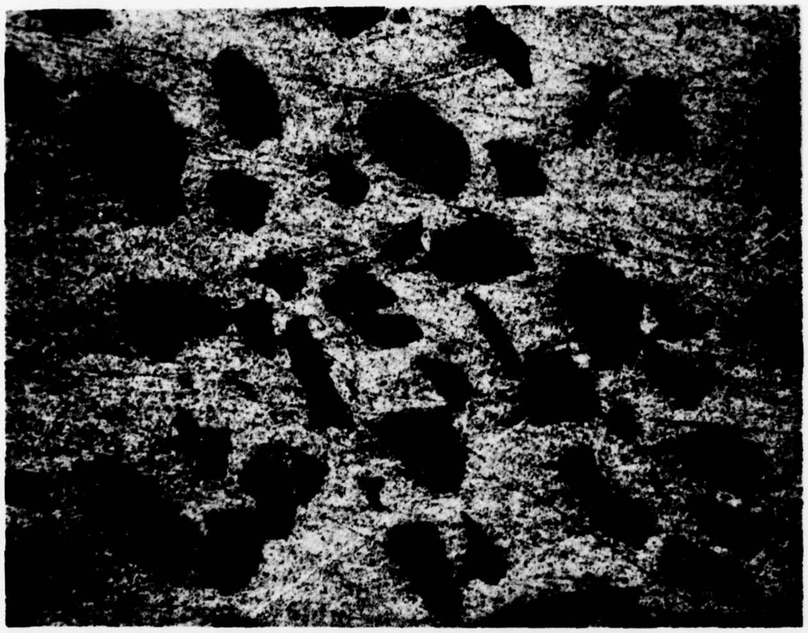
(c) 750X

FIGURE 8 - Specimen current scanning electron micrographs of three particles in Series 2 hot pressing. Electron probe analyses of the indicated points are listed in Table V.



P.N. 1-74055 250X

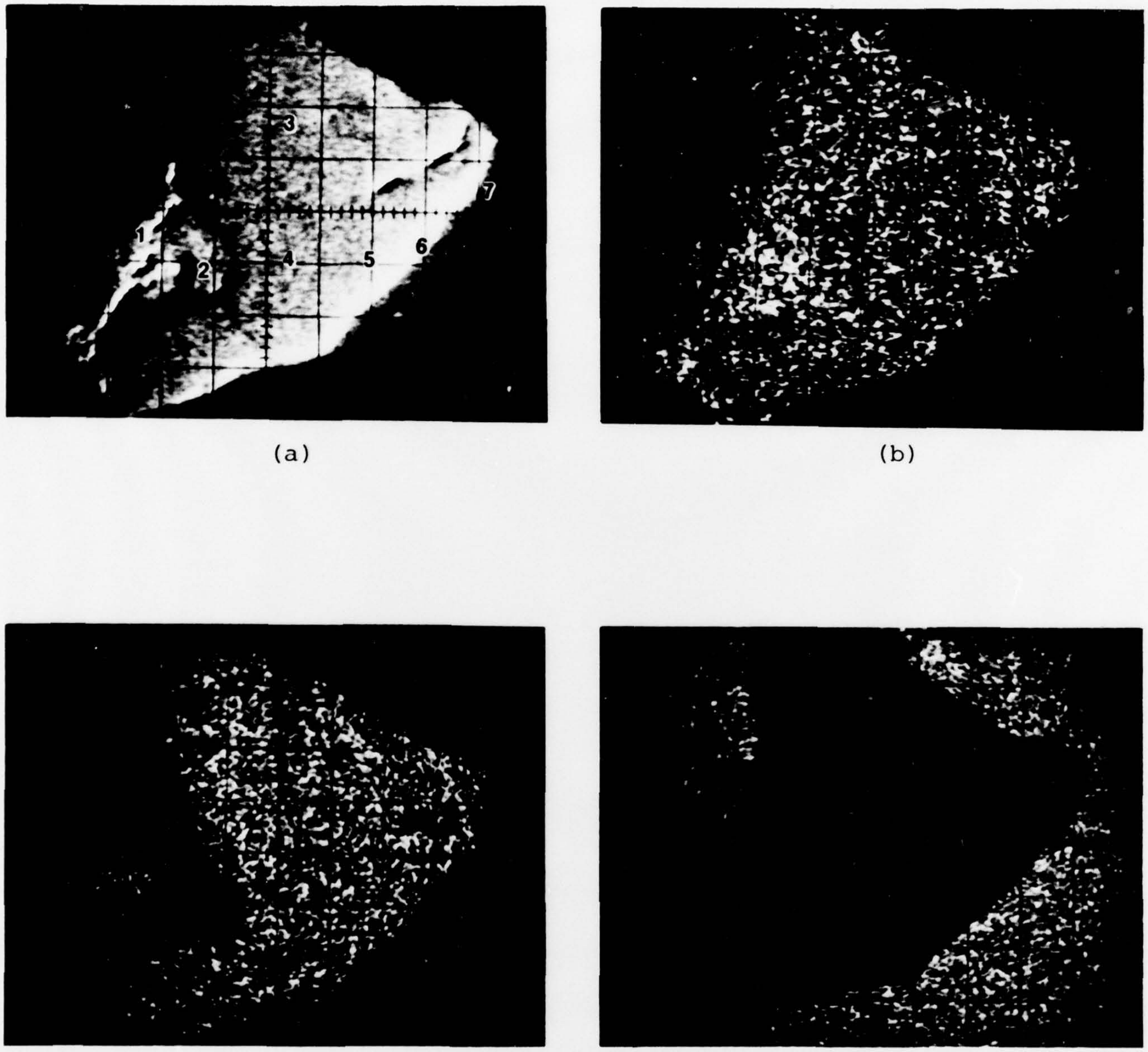
FIGURE 9 - Optical micrographs of Sm-Dy-Fe alloy, T-82138, in the as-cast condition. Etched in 2% Nital.



P.N. 1-75078 (a) 100X

P.N. 1-75090 (b) 500X

<u>FIGURE 10</u> - Optical photomicrographs of Series 3 $Sm_{0.88}Dy_{0.12}Fe_2$/Ni composites as hot pressed at 1200°F. Final polish with 1μ diamond. Unetched.



(a) (b)

(c) (d)

FIGURE 11 - (a) Specimen current SEM of typical particle of Series 3 1200°F hot pressing. (b) Ni K_α image. (c) Fe K_α image. (c) Sm L_α image. Magnification = 500X.

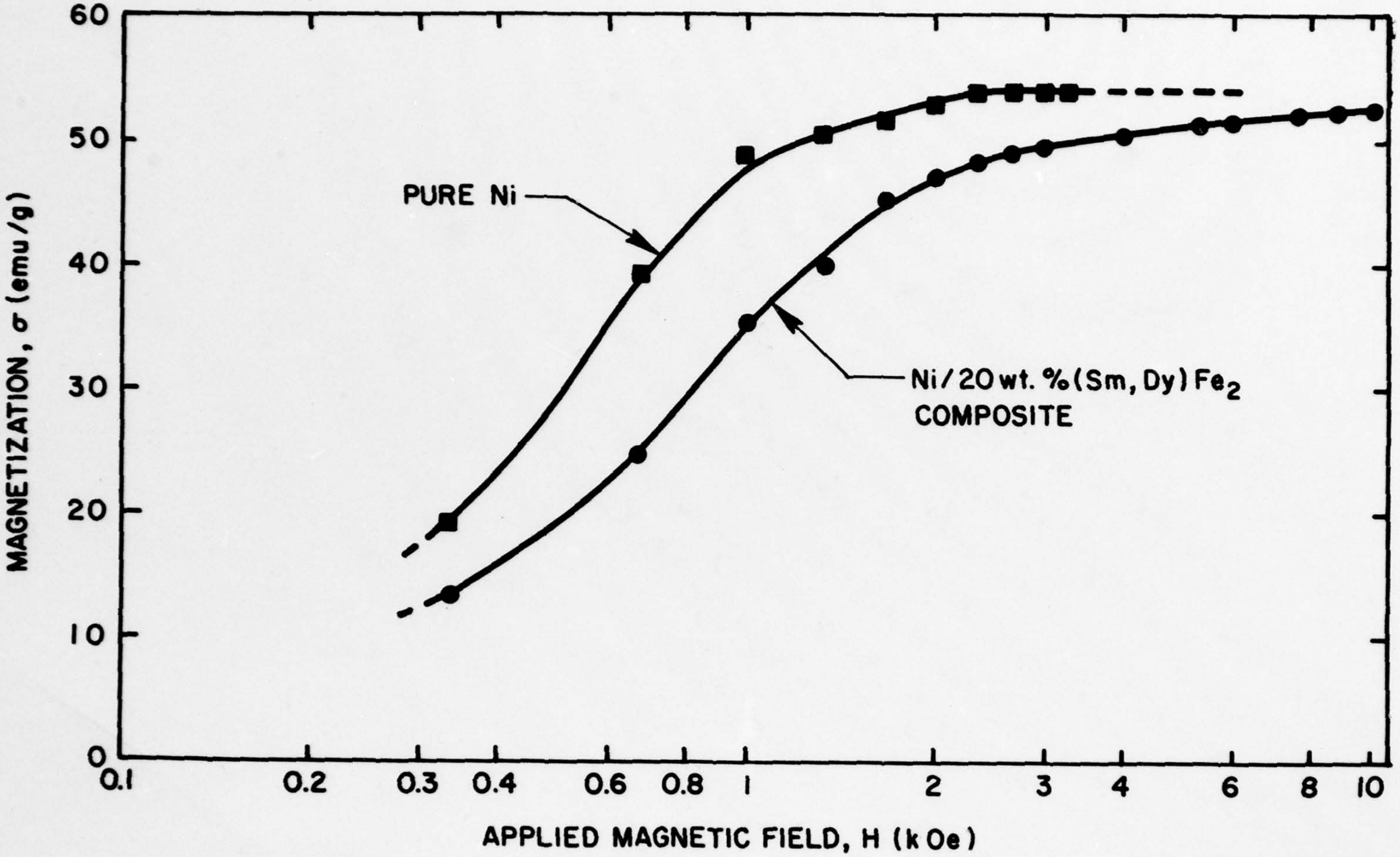


FIGURE 12 - MAGNETIZATION AS A FUNCTION OF APPLIED FIELD FOR THE SERIES 3 1200°F (649°C) HOT PRESSED Ni AND Ni/20 wt.% (Sm, Dy) Fe_2 SPECIMENS.