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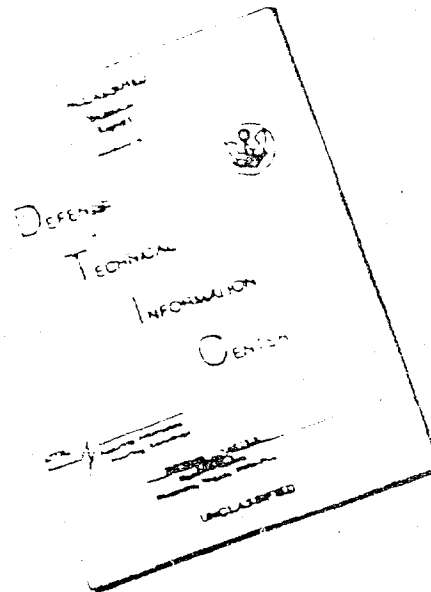
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The Preparation and
Characteristics of

THIN FERROMAGNETIC FILMS

SCIENTIFIC REPORT NO. 1
CONTRACT AF19(604)-4978

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MAY 1960

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Characteristics of
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Prepared for
Electronics Research Directorate
Air Force Cambridge Research Center
Air Research and Development Command
United States Air Force
Bedford, Massachusetts

Remington Rand Univac[®]
DIVISION OF SPERRY RAND CORPORATION
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ACKNOWLEDGEMENTS

The developments described in this report were carried out in the Materials Development Department under the management of Dr. Charles J. Kriessman. Many of the detailed experiments were performed in the Materials Engineering Department under the supervision of Dr. Joseph Mathias, who was also directly responsible for much of the electroplating research.

Dr. Arnold Schmeckenbecher directed the chemical decomposition studies and Mr. Edward Schneider worked on the electroplating of thin films. All physical measurements were carried out under the direction of Mr. Alfred Adomines.

The authors gratefully acknowledge the suggestions given by Dr. Henry Belson and Mr. Thomas Matcovich, both of the Solid State Physics Laboratory.

Technicians who contributed greatly to the success of the project are Charles Bell, Guy DiGuillio, Edward Kostyk, Samuel Mason, and John Tichy.

ABSTRACT

This report describes the results of an experimental study of the magnetic properties of thin films prepared by chemical methods. Electrodeposition in aqueous solutions and thermal decomposition of carbonyl vapors have shown very encouraging results.

Electrodeposited ferromagnetic nickel-iron films having thicknesses in the range 2500 to 3000Å give reproducible results when a glass substrate is used onto which a conductive layer of gold is sputtered. Films deposited on Mylar having a conductive layer of evaporated chromium and copper also give good magnetic properties. Such films exhibit an average coercive force of about 1.2 oersteds and H_k on 3 to 5 oersteds depending on their composition. Switching constants of 0.05 to 0.20 oersted-microsecond are obtained by application of crossfields of about 0.25 H_k .

Films produced by thermal decomposition of carbonyl mixtures do not as yet show comparable magnetic or switching properties. Control of this process is quite difficult, but fairly reproducible results can be obtained by depositing the films on glass substrates and subsequently annealing them in a hydrogen atmosphere. The average values of coercive force obtained after annealing for films of about 3000Å are 2.5 oersteds.

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SECTION 1

INTRODUCTION

This is a report on work carried out under Contract AF-19(604)-4978 sponsored by the Air Force Cambridge Research Center for the period January 1, 1959 to February 1, 1960 and is an extension of Contract AF-19 (604)-2659 which covered the period August 1, 1957 to August 31, 1958. A substantial portion of the work discussed herein was supported by internal funds of the Remington Rand Univac[®] Division of the Sperry Rand Corporation.

The objectives of this program are to prepare thin ferromagnetic films by chemical techniques, to determine the effect of variations in processing procedures on the properties of the films, and to study experimentally and theoretically the physical and magnetic properties of the films.

Emphasis has been placed on the investigation of the usefulness of chemical techniques for a number of reasons. One of these is the fact that extensive research is being carried out by a number of laboratories on the vacuum evaporation method for preparing thin films. Also, vacuum evaporation has certain limitations which include (1) the lack of high reproducibility of film properties such as the ratio of the coercive force to the anisotropy field (H_c/H_k) and the anisotropy direction needed for application in high speed computers, and (2) the inability to produce large arrays of films over large areas. Chemical methods eventually may yield better reproducibility and larger arrays. Also, the use of the elaborate equipment necessary for the production of a high vacuum makes this procedure inherently more expensive than the chemical processes.

During the course of this work, all chemical techniques useful in the preparation of thin ferromagnetic films have been eliminated except two: electrodeposition and thermal decomposition of carbonyls. The previous report on this work described the results of preliminary investigations on the production of thin films by the thermal decomposition of acetylacetonates and by electrodeposition. Several of the films produced showed fairly good magnetic properties but all of them had a high switching constant and did not compare favorably with vacuum deposited films. Reproducibility of the films also was a problem. Consequently, a major portion of the work carried out under the present contract was directed towards obtaining reproducible films with improved switching characteristics.

The thermal decomposition of mixtures of nickel and iron carbonyl also was investigated as a means of producing ferromagnetic thin films.

During this period the condition for obtaining reproducible films by the electrodeposition technique have been determined. One of the most important conditions for reproducibility is the choice of a substrate. Solid metal substrates give universally poor results, while gold films sputtered on glass and chromium-copper films evaporated on Mylar give good results.

In 40 separate electrodepositions of sputtered gold substrates the values of H_c have been held constant within ± 15 percent for films which were 2500 Ångstroms thick. These films had switching constants of 0.05 oersted-microsecond when crossfields of less than one quarter the anisotropy field (H_k) were applied. Films produced by thermal decomposition of carbonyls do not as yet show magnetic or switching properties comparable to those obtained from vacuum deposited or electroplated films. However, under special processing conditions, films are produced which have unique properties termed "threshold" properties. These properties suggest new uses for magnetic films in computer circuits.

SECTION 2

ELECTRODEPOSITION OF NICKEL-IRON FILMS

2-1. INTRODUCTION

In a previous report the principles of electrodeposition of alloys and the factors that could theoretically affect the properties of films formed by this technique were discussed. Therefore, this section will deal primarily with the experimental work carried out during this report period.

For purposes of electrodeposition it is necessary to have a conductive substrate. Since the films deposited are very thin, the substrate and the quality of its surface have a major effect on the magnetic properties of the films deposited on it. For this reason various substrates have been tried in order to evaluate their usefulness. Another major area of investigation has been the plating conditions. The factors investigated are the current density and the composition of the solution.

2-2. PLATING EQUIPMENT

The plating cell consists of a lucite plastic chamber 6 inches long, 3 inches wide and 3 inches high. A cross section of the cell is shown in figure 2-1 and a photograph of the equipment in figure 2-2. The contents of the cell are continuously filtered by pumping the solution through a five-micron fritted-glass filter set into a large test tube. The large test tube is immersed in a water bath which can be heated by means of an immersion heater. In this way the temperature of the plating bath can be raised to the desired level and maintained constant by a temperature sensing element immersed in the plating bath which controls the input power to the heater in the water bath. The temperature of the solution can be maintained constant within $\pm 2^{\circ}\text{C}$. Current is supplied from a 6-volt battery and an ammeter and voltmeter are incorporated in the circuit. Plating is done in the presence of a linear magnetic field which is provided by a large permanent magnet. This magnet has a 1-inch pole face and is placed so that the faces are against the two sides of the cell and the centers of the faces are in line with the center of the 9mm specimen being plated. The magnetic field created by the magnet was found to be 400 oersteds at the center of a gap of 3-1/2 inches.

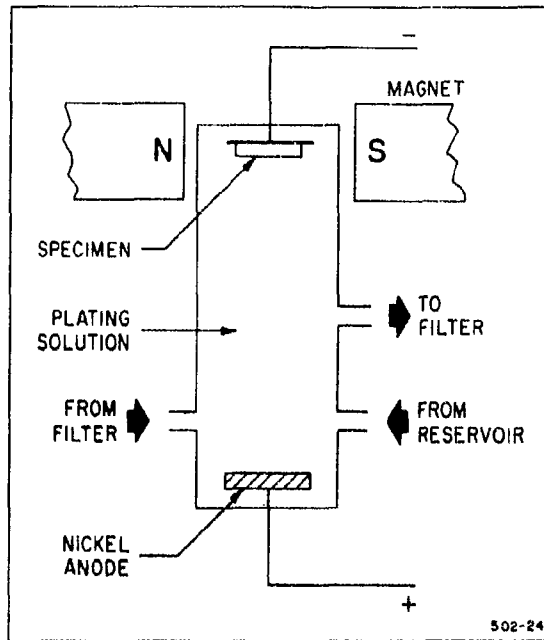


Figure 2-1. Outline of a Plating Cell

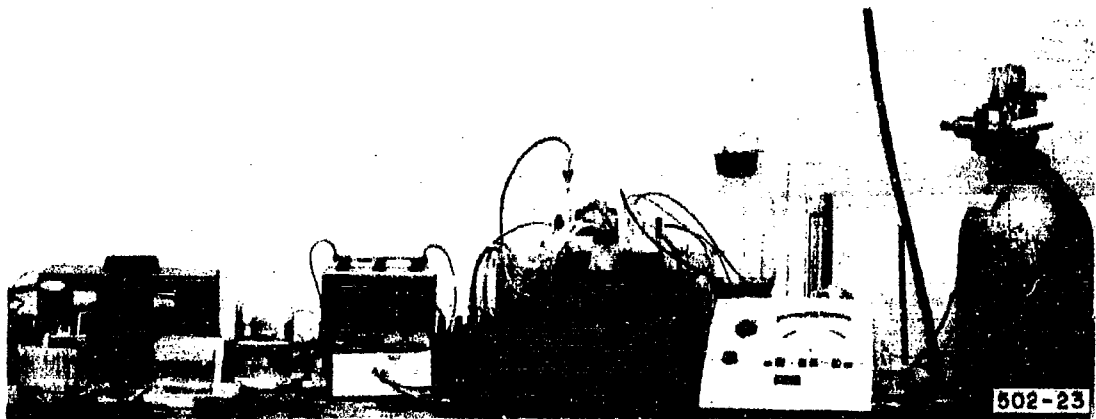


Figure 2-2. Apparatus for Electrodeposition of Nickel-Iron Films

**Table 2-1. Magnetic Properties of Thin Films
Electrodeposited on Various Substrates**

Temperature: 25°C Current density: 6 ma/cm²
 Field: none Average thickness: 3000Å
 Plating time: 240 seconds Average composition: 20% Fe - 80% Ni

All data taken at 10-oersted drive
 All data are average of 3 runs

Substrate	Magnetic Properties					Series (PI-)
	Easy Direction		Hard Direction		Anisotropy Field	
	H _c (oe)	B _r /B _m	H _c (oe)	B _r /B _m	H _k (oe)	
Mechanically polished bulk brass	3.8	0.82	1.8	0.43	6	291
Mechanically polished bulk copper	3.5	0.85	2.2	0.67	5-6	291
Rolled silver on aluminum sheet	3.0	0.83	2.0	0.55	6-7	291
Rolled gold on aluminum sheet	4.3	0.90	2.0	0.60	6-7	289
Chromium-copper evaporated on nylon	1.4	0.90	0.7	0.23	4	290
Chromium-copper evaporated on Mylar	1.6	0.95	1.1	0.36	4	301
Gold sputtered on glass	1.4	0.95	0.8	0.30	4-5	297

2-3. EFFECT OF SUBSTRATES

Several series of experiments were carried out under identical conditions using a variety of substrates. Typical data obtained are shown in table 2-1. Films electroplated on bulk metal substrates have relatively high H_c, low B_r to B_m ratios, and high values of H_k. Reproducibility of these values from substrate to substrate also is very poor. This probably is due to the variations in grain size and surface roughness and strains that always are present in metals. When the substrate does not have a recognizable grain structure as in thin sputtered or evaporated films, the reproducibility of the magnetic properties under identical plating conditions is good. Table 2-2 contains typical results obtained from several plating runs using gold sputtered on cover-glass slides and chromium-copper evaporated onto 1-mil Mylar* film as substrates. Chromium is used on the Mylar only to obtain good adherence.

* Registered trademark, E. I. du Pont de Nemours Company, Wilmington, Del.

Table 2-2. Magnetic Properties of Thin Films Using Gold Sputtered on Glass and Chromium-Copper Evaporated on Mylar as Substrates

Temperature: 52°C Current density: 6 ma/cm²
 Field: 400 oersteds Average thickness: 3000Å
 Plating time: 240 seconds Average composition: 20% Fe, 80% Ni

All data taken at 10-oersted drive

Series (PI-)	Magnetic Properties				
	Easy Direction		Hard Direction		Anisotropy Field
	H _c (oe)	B _r /B _m	H _c (oe)	B _r /B _m	H _k (oe)
Substrate: Gold Sputtered on Glass					
277/6	1.2	0.95	1.2	0.36	5.0
277/7	1.0	0.95	0.7	0.23	5.4
277/8	1.3	0.95	0.7	0.31	5.0
277/12	0.9	0.97	0.7	0.29	4.5
283/10	0.8	0.92	0.6	0.20	4.0
283/11	1.5	0.96	0.9	0.35	4.0
283/12	1.5	0.97	0.8	0.30	4.0
283/13	1.3	0.96	0.7	0.24	4.0
Substrate: Chromium-Copper Evaporated on Mylar					
284/2	1.0	0.94	0.65	0.20	4.5
284/9	1.8	0.95	0.60	0.31	4.5
284/10	1.5	0.91	0.90	0.18	4.0
284/11	2.0	0.94	0.58	0.33	4.0
284/14	1.4	0.95	0.50	0.20	4.5

Table 2-3. Effect of Current Density on Magnetic Properties, I

Substrate: Gold sputtered on glass Average thickness: 1500Å
 Temperature: 25°C Series: PI-274
 Average composition: 20% Fe - 80% Ni

All data taken at 10-oersted drive
 All data are average of 3 runs

Current Density (ma/cm ²)	Time (sec)	Magnetic Properties				
		Easy Direction		Hard Direction		Anisotropy Field
		H _c (oe)	B _r /B _m	H _c (oe)	B _r /B _m	H _k (oe)
25.00	30	6.1	0.94	7.3	0.92	5
12.50	60	5.0	0.92	3.7	0.92	5
6.25	120	3.6	0.92	3.1	0.92	4
3.10	240	10.5	0.86	7.5	0.92	6

Table 2-4. Effect of Current Density on Magnetic Properties, II

Temperature: 25°C

All data taken at 10-oersted drive

All data are average of 3 runs

Current Density (ma/cm)	Iron Content (percent)	Thickness (Å)	Magnetic Properties				
			Easy Direction		Hard Direction		Anisotropy Field
			H _c (oe)	B _r /B _m	H _c (oe)	B _r /B _m	H _k (oe)
Substrate: Gold Sputtered on Glass							
3	24.2	1650	5.8	0.97	3.0	0.60	6.0
4	23.1	2180	4.1	0.97	3.1	0.70	6.0
5	23.0	2260	3.7	0.98	2.7	0.70	6.0
6	23.0	2050	1.8	0.97	1.4	0.50	4.4
3	18.7	2130	3.4	0.97	1.5	0.37	5.0
4	22.5	2020	2.6	0.95	1.4	0.28	4.0
5	20.7	2840	1.5	0.97	1.2	0.40	5.0
6	20.5	2540	1.3	0.96	1.1	0.37	4.0
4	—	2400	1.6	0.95	1.1	0.24	4.2
5	18.2	2510	1.6	0.95	1.6	0.49	5.0
6	20.4	3070	1.1	0.96	0.8	0.31	4.1
Substrate: Chromium-Copper on Mylar							
3	—	—	4.8	0.96	2.9	0.73	10.0
4	—	—	2.9	0.96	1.6	0.60	5.0
5	22.7	1780	2.9	0.97	1.7	0.49	5.0
6	22.6	1930	3.4	0.98	2.1	0.69	5.5
3	—	2340	3.5	0.96	2.3	0.56	8.0
4	23.4	2490	2.4	0.94	1.3	0.55	5.0
5	—	—	1.8	0.95	1.1	0.40	5.0
6	23.5	2340	1.8	0.94	1.2	0.45	3.6
3	21.3	2570	3.0	0.96	1.6	0.46	5.7
4	18.8	—	1.8	0.95	1.9	0.31	5.5
5	23.8	3190	1.0	0.90	0.7	0.23	4.0
6	19.6	2950	1.6	0.94	0.7	0.30	4.3

2-4. EFFECT OF CURRENT DENSITY

In all electrodeposition processes there is a certain range of current density in which the best coatings can be obtained. The criteria for good electrodeposits in decorative plating are adhesion, brightness, lack of pin holes, and so forth. For economic reasons the maximum current density compatible with the above criteria is used in practice. In the electrodeposition of magnetic films it appears that the rate of deposition of the film and consequently the current density of electrolysis have consider-

Table 2-5. Composition of the Plating Bath

Chemical	Formula	Quantity (grams/liter)
Nickel (II) Sulfate	$\text{NiSO}_4 \cdot 6\text{H}_2\text{O}$	218.00
Sodium Chloride	NaCl	9.70
Boric Acid	H_3BO_3	25.00
Saccharin	$\text{C}_6\text{H}_4\text{SO}_2\text{NHCO}$	0.83
Triton CF-21	(Trademark of Rohm & Haas Co. Phila., Pa.)	1.30

Table 2-6. Effect of Variation of Ferrous Iron Content in the Plating Solution on Magnetic Properties

Substrate: Gold sputtered on glass

Current density: 6 ma/cm²

All data taken at 10-oersted drive

Series (PI-)	Iron Content (percent)	Thickness (Å)	Magnetic Properties				
			Easy Direction		Hard Direction		Anisotropy Field
			H _c (oe)	B _r /B _m	H _c (oe)	B _r /B _m	
286/3	22.5	2020	2.0	0.96	0.90	0.40	4.5
286/4	—	—	1.5	0.95	0.88	0.30	4.5
286/5	—	—	2.0	0.96	1.00	0.96	4.5
296/6	—	—	1.9	0.96	1.00	0.35	4.5
297/15	18.1	—	—	—	—	—	—
297/16	—	—	1.0	0.91	0.70	0.23	2.6
297/17	—	—	0.9	0.98	0.70	0.20	3.5
298/1	—	3000	0.7	0.96	0.80	0.23	3.5
298/2	—	—	1.0	0.91	1.00	0.30	3.4

able influence on the magnetic properties. Since thickness of the deposit also has an influence on the magnetic properties, a survey of the current density range from 3 to 25 ma/cm² at a thickness of 1500 Ångstroms first was made using gold sputtered on cover-glass slides as a substrate. This data shown in table 2-3, indicates that a current density of about 6 ma/cm² produced the best properties. Further experiments in the more limited range of 3 to 6 ma/cm² showed that 5 and 6 ma/cm² gave the best properties in the thickness range 2000 to 3000 Ångstroms. The results of these experiments are shown in table 2-4 for the two substrates, gold sputtered on glass and chromium-copper evaporated on Mylar.

2-5. COMPOSITION OF PLATING SOLUTION

For use in the new plating apparatus, three-liter quantities of plating solution were prepared and kept under an atmosphere of nitrogen to prevent oxidation of the ferrous iron. The formula of the solution shown in table 2-5 is similar to that used in the previous work except that the salt supplying a common ion, magnesium sulfate, is not included. A small amount of sodium chloride is included since the presence of chloride ions is necessary to render the nickel anode active in the electroplating process. The circulating action of the present plating cell causes the sodium luryl sulfate to foam excessively and it was therefore replaced with a nonfoaming wetting agent, Triton CF-21.* Only pure reagent chemicals were used.

Varying amounts of ferrous sulfate ($\text{Fe SO}_4 \cdot 7\text{H}_2\text{O}$) were added to the solution shown in table 2-5 while the nickel content was kept constant. Thus, to prepare nickel-iron films of 80:20 composition at 3000 Ångstroms thickness, 6 grams of ferrous sulfate per liter were added.

Plating was conducted at pH range of 2.9 to 3.0. Adjustments in pH were made by addition of dilute sodium hydroxide (5 percent) or sulfuric acid (1:4). Similarly, a plating solution containing less iron (4.8 grams ferrous sulfate per liter) was used in order to prepare nickel-iron films with a composition of approximately 18 percent iron at 3000 Ångstroms.

Table 2-6 contains data for magnetic properties of films plated from solutions with two different ferrous iron concentrations. Decreased iron content results in lower values of H_k . The same result has been observed in evaporated films. Films with lower iron content also have a slightly lower H_c .

2-6. EFFECT OF FILM THICKNESS ON MAGNETIC PROPERTIES

Thin films were plated on gold sputtered on glass substrates at a current density of 6 ma/cm^2 for different time periods to produce films within a range of approximately 1000 to 3000 Ångstroms thickness.

Data presented in table 2-7 show the results obtained on film deposited from solutions of 20 and 18 percent iron content (6.0 and 4.8 gms/liter $\text{Fe SO}_4 \cdot 7\text{H}_2\text{O}$).

The differences shown by the two sets of data are very slight. The one significant difference is the tendency for lower H_k values in films of lower iron content.

The general tendency for the values of H_c and H_k to decrease as thickness increases is evident in both compositions.

* Trademark of Rohm and Haas Co., Phila., Pa.

Table 2-7. Effect of Thickness on Magnetic Properties

Substrate: Gold sputtered on glass
Current density: 6 ma/cm²

All data taken at 10-oersted drive

Series (PI-)	Time (sec)	Iron Content (percent)	Thickness (Å)	Magnetic Properties				
				Easy Direction		Hard Direction		Anisotropy Field
				H _c (oe)	B _r /B _m	H _c (oe)	B _r /B _m	H _k (oe)
Solution A: Nominal Iron Content 20%								
281	75	25.2	1000	6.3	0.94	4.8	0.78	7
—	150	22.6	2400	3.3	0.98	1.7	0.57	6
286	180	22.5	2600	1.5	0.95	1.1	0.35	4
281	210	22.0	2800	1.1	0.94	0.7	0.30	4
283	240	21.9	3150	1.2	0.96	0.7	0.25	4
281	270	20.0	3600	1.0	0.92	0.9	0.32	4
Solution B: Nominal Iron Content 18%								
305	75	—	1000	5.5	0.94	4.2	0.78	7
300	120	20.5	1500	4.4	0.96	2.7	0.68	6
300	150	23.0	1880	2.6	0.95	1.7	0.45	5
300	180	18.3	2450	1.4	0.93	1.0	0.30	4
297	210	19.7	2710	1.2	0.95	0.8	0.27	3-4
297	240	19.0	3070	1.3	0.95	1.1	0.34	3-4

2-7. SWITCHING PROPERTIES OF ELECTRODEPOSITED FILMS

Several films have been tested on the transmission line switching apparatus shown in block diagram form in figure 2-3. A picture of the actual experimental apparatus is shown in figure 2-4. Figures 2-5 and 2-6 show some examples of the switching curves obtained from two films at various crossfields. It is noticed that at crossfields close to the anisotropy field the remagnetization process is so rapid that it cannot be measured by the present equipment.

2-8. CONCLUSIONS

The results of this investigation into the preparation of thin films by electroplating are the following:

(1) The conditions for obtaining reproducible films having a thickness of 2500 Ångstroms, H_c = 1.0 oersted and B_r/B_m of 0.95 have been developed.

(2) Gold sputtered on glass and evaporated chromium-copper on Mylar have proved to be the best substrates for obtaining reproducible films.

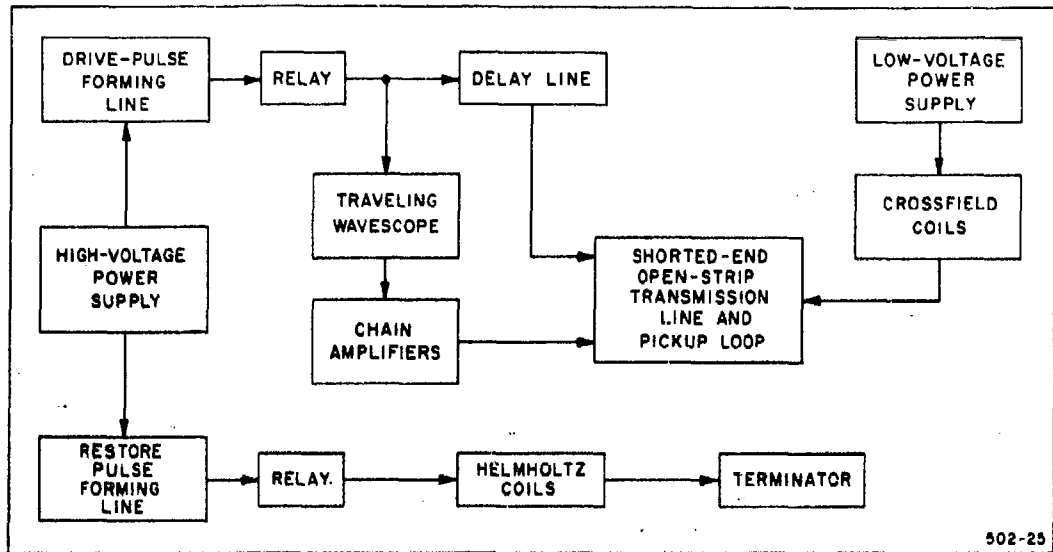


Figure 2-3. Block Diagram, Apparatus for High-Speed Switching of Thin Nickel-Iron Films

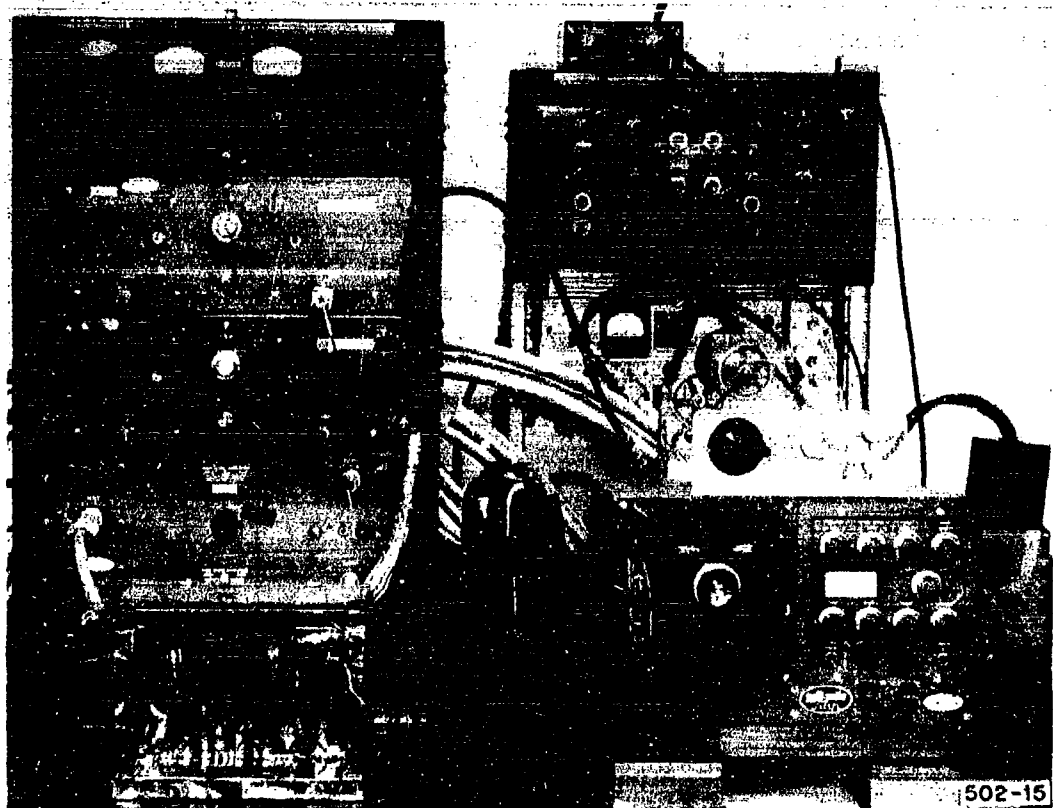


Figure 2-4. Photo, Apparatus for High-Speed Switching of Thin Nickel-Iron Films

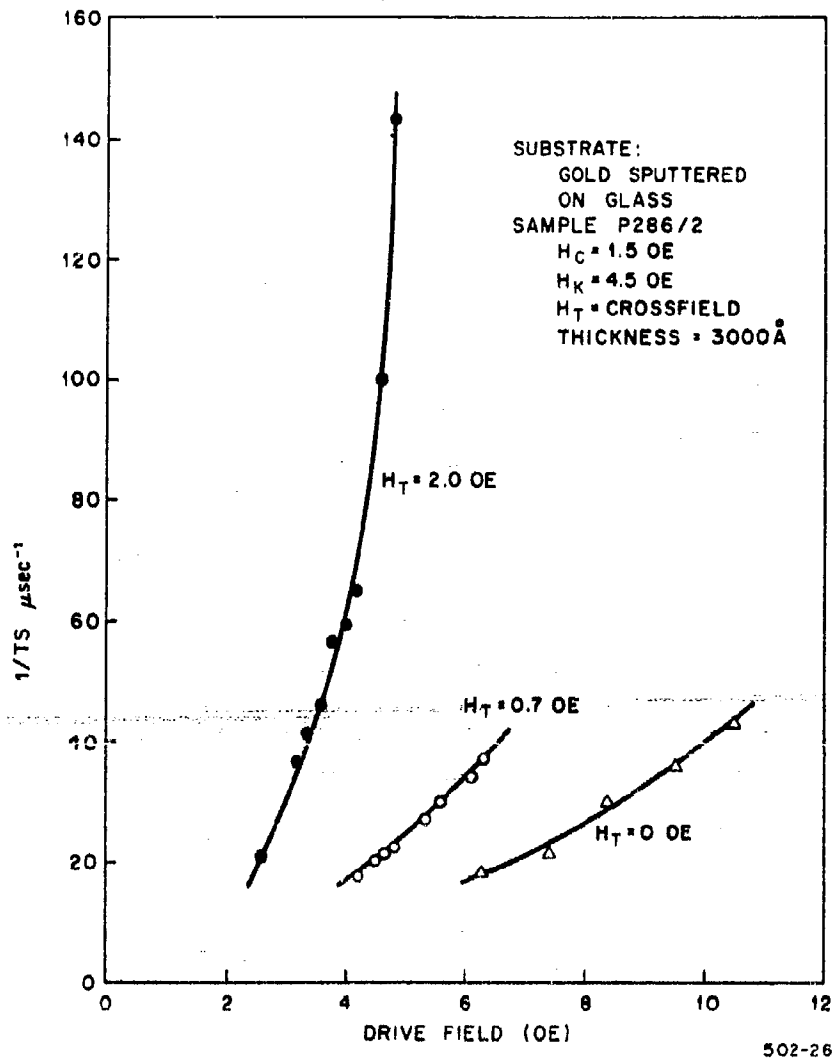


Figure 2-5. Switching-Time Curve I
 for Electrodeposited Film

(3) The values of H_k are dependent on the composition of the film thickness. Values of H_k in the range of 3 to 5 oersteds are obtained.

(4) Switching constants of electroplated films are as good as those obtained from vacuum deposited films. Switching constants of 0.05 oersted-microsecond and better can be obtained with crossfields of less than $0.25 H_k$.

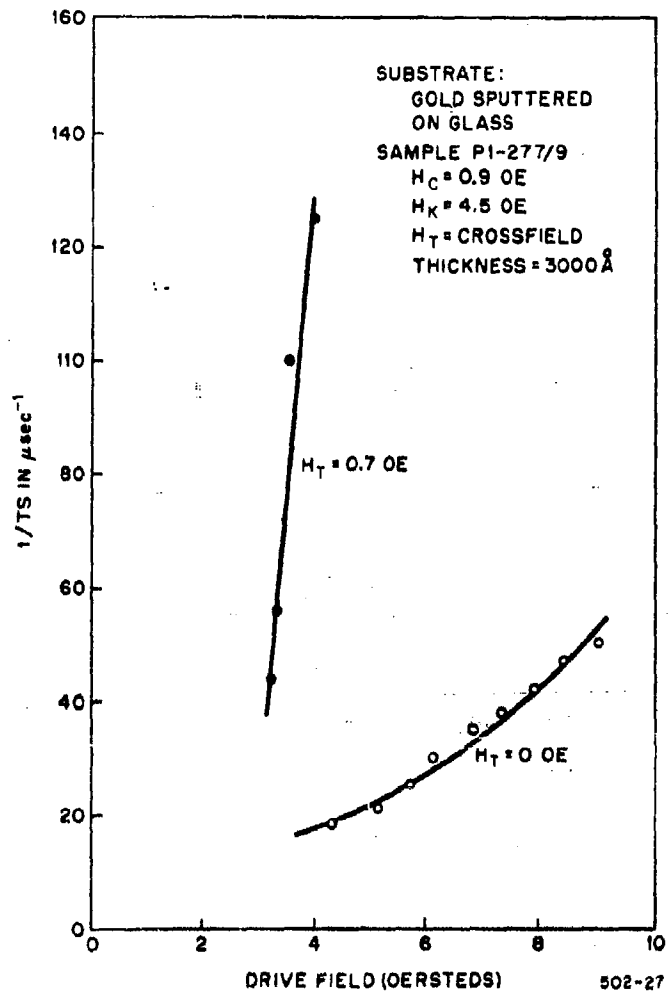


Figure 2-6. Switching-Time Curve II for Electrodeposited Film

SECTION 3

FORMATION OF NICKEL-IRON FILMS BY THERMAL DECOMPOSITION OF ACETYLACETONATES

3-1. INTRODUCTION

Previous work in preparing thin nickel-iron films by thermal decomposition of acetylacetonates indicated that this technique produced films with high coercive force and low switching constant even after annealing. Reproducibility and control of composition were the major problems in using this method. During this report period attempts were made to improve the control over the composition by controlling vapor pressures of the metal acetylacetonates, and by process changes. However, it was decided at a later date to drop further work in acetylacetonates and to concentrate on carbonyls for the following reasons:

- (1) The vapor pressure of nickel acetylacetonates cannot be controlled well because of the formation of glassy polymers.
- (2) The vapor pressure of the metal acetylacetonates is very low at room temperature; therefore, to prevent condensation all of the equipment would have to be maintained at elevated temperatures.
- (3) The decomposition of metal carbonyls is by far an easier and more controllable process than the decomposition of metal acetylacetonates.

3-2. VAPOR PRESSURE-TEMPERATURE RELATIONSHIP OF IRON AND NICKEL ACETYLACETONATES

In order to control the composition of the film obtained by the decomposition of acetylacetonates it is necessary to know the relationship between vapor pressure and temperature of these compounds. Since this data was not available in the literature, equipment shown in figure 3-1 was designed to obtain it.

A stream of hydrogen was passed over a sample of vacuum-sublimed iron acetylacetonate kept at 170°C (308°F). The vapor then was passed through an area kept at one of three temperatures: 130°C (234°F), 140°C (252°F), or 150°C (270°F). Some of the iron acetylacetonate condensed in this region. The hydrogen, now saturated at 130°C (or at 140°C or 150°C), then was passed through a zone kept at room temperature (27 to 29°C, 80.6 to 84.2°F).

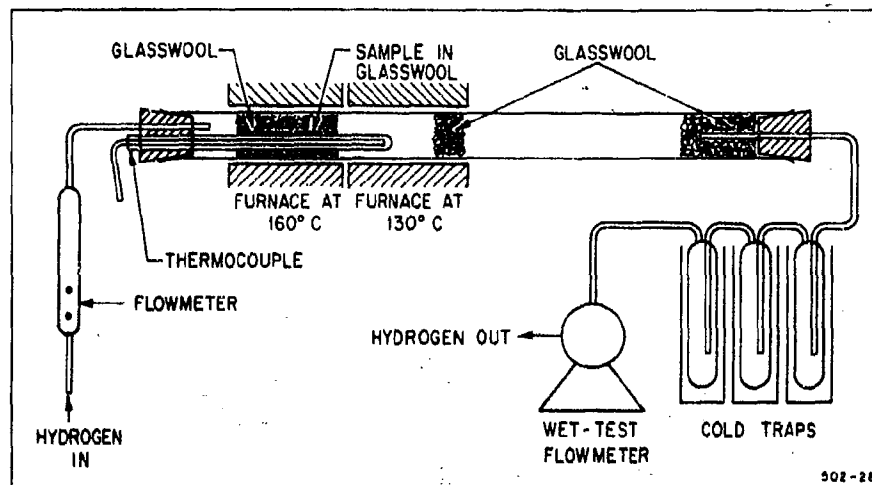


Figure 3-1. Apparatus for Measuring Vapor Pressure of Ferric Acetylacetonates

The amount of iron acetylacetonate condensed in this zone was determined by decomposing the iron acetylacetonate in acid and determining the iron content of the acid solution. The amount of hydrogen passed through the system during the experiment was measured by a wet-test flowmeter. The vapor pressure data obtained is shown in figure 3-2. This data can be fitted to the following equation:

$$\text{Log } p = 17.14 - \frac{6300}{T}$$

where p is in microns Hg and T is in degrees Kelvin. No data for the vapor pressure of nickel acetylacetonate can be obtained by the method used for the determination of the vapor pressure of ferric acetylacetonate because it is necessary to saturate the hydrogen carrier gas at a temperature close to the decomposition temperature (190 to 195°C, 362 to 371°F) of the acetylacetonate. Some of the nickel acetylacetonate decomposes and tends to dissolve in the undecomposed material.

In the course of some of the experiments, the solvent was re-evaporated and the nickel acetylacetonate that was left appeared like a "glass". The vapor pressure of this material was found to be practically zero at 190°C.

The apparatus shown in figures 3-3 and 3-4 was constructed to mix separate streams of hydrogen saturated with the two different acetylacetonates and to decompose the mixture in an adjacent chamber held at 390°C (702°F). Several experiments were carried out with this apparatus, but the resultant films contained practically no nickel because of the formation of glassy polymers in the nickel tube.

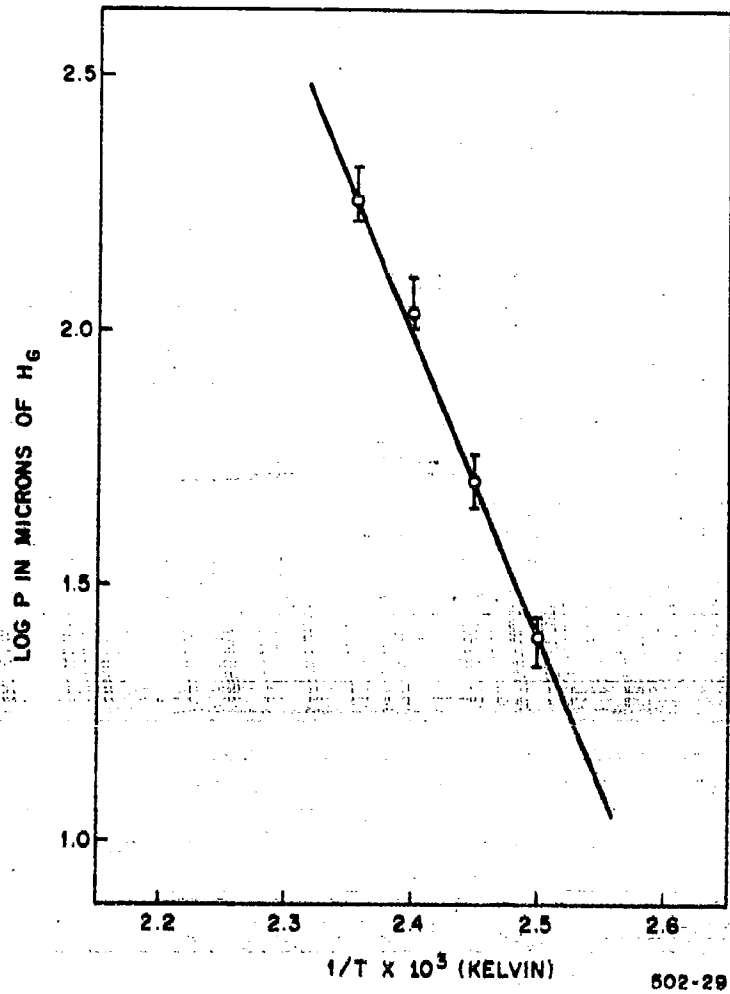


Figure 3-2. Vapor Pressure-Temperature Relationship of Ferric Acetylacetonates

3-3. CONCLUSIONS

Because of the difficulty in maintaining a satisfactory concentration of nickel acetylacetonate in a hydrogen stream, it appears that this method cannot be used to obtain useful nickel-iron thin films. Iron films can be produced easily by this method. Nickel-iron films could be obtained if a method were found to prevent the decomposition and polymerization of the nickel acetylacetonates. This polymerization of the acetylacetonates into a glassy phase might be useful in the future for preparing other types of magnetic films such as ferrite films.

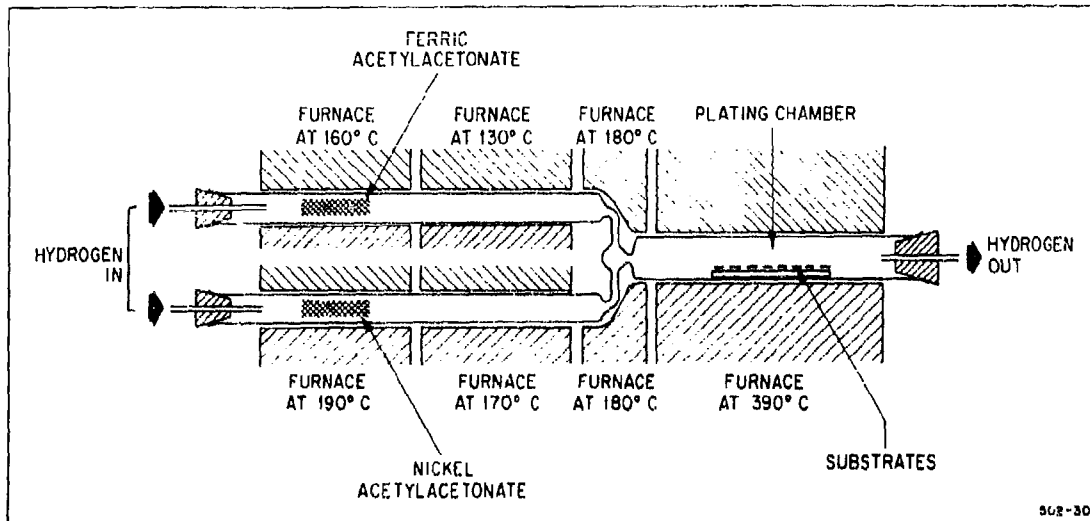


Figure 3-3. Diagram, Apparatus for Making Nickel-Iron Films by Decomposing Metal Acetylacetonates

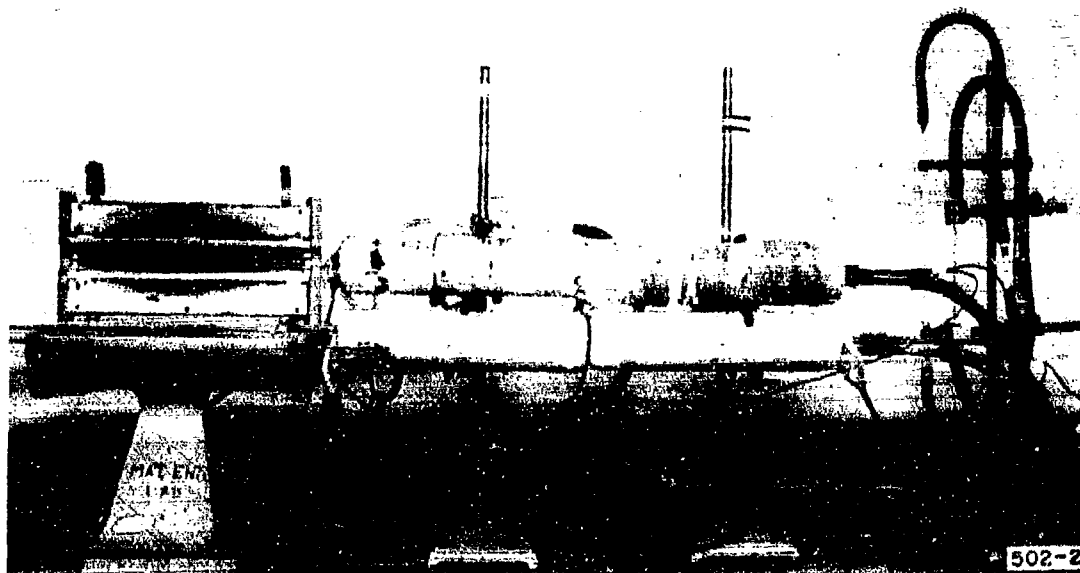


Figure 3-4. Photo, Apparatus for Making Nickel-Iron Films by Decomposing Metal Acetylacetonates

SECTION 4

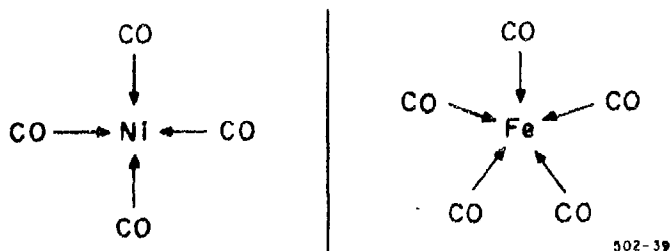
FORMATION OF NICKEL-IRON FILMS BY THERMAL DECOMPOSITION OF IRON AND NICKEL CARBONYLS

4-1. INTRODUCTION

The pyrolysis of nickel carbonyls to produce a high grade of nickel is an industrial process that has been used for many years.^{2,3,4} Apparatus for continuously coating metal strips⁵ and for coating small objects on an assembly line basis⁶ has been developed and is being used commercially. Iron carbonyl also has been used in the production of iron powder for powder metallurgical preparation of magnetic cores. In most cases the iron produced by this method has a high coercivity and requires extended annealing in a hydrogen atmosphere to remove the carbon. The use of the pyrolysis of the carbonyls for the preparation of magnetic thin films thus far has not been reported in the literature. However, the easy availability of these materials and their high vapor pressure appear to make this process quite attractive. For these reasons, investigations into the types of films that could be produced from the pyrolysis of carbonyls were started in this report period with encouraging results.

4-2. THEORETICAL CONSIDERATIONS

Nickel tetracarbonyl $[\text{Ni}(\text{CO})_4]$ has the four carbon-monoxide groups arranged around the central nickel atom. Each CO group shares one electron pair with the nickel atom filling in the 3d, and 4p orbits to obtain the stable configuration of the krypton atom. Similarly, the five carbonyl groups in the iron pentacarbonyl $[\text{Fe}(\text{CO})_5]$ brings the number of electrons around the central iron atom to the stable krypton configuration.



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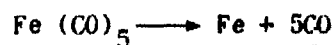
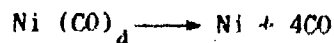
As a result of the noble gas configuration of the carbonyl molecule the intermolecular forces are relatively weak, resulting in an unusually high vapor pressure. Nickel carbonyl melts at -25°C (-13°F) and boils at 43°C (77.4°F). The vapor pressure of the liquid is given by the equation⁷

$$\text{Log } p = 7.878 - \frac{1574.49}{T}$$

At 25°C (77°F) the vapor pressure is 394mm of Hg. Iron pentacarbonyl melts at -21°C (-5.8°F) and boils at 104°C (219.2°F). At 25°C its vapor pressure is 26.8mm Hg and the vapor pressure of the liquid pentacarbonyl is given by the equation⁷

$$\text{Log } p = 8.3098 - \frac{2050.7}{T}$$

Both nickel and iron carbonyls decompose with the deposition of the metal and liberation of carbon monoxide according to the equations



The decomposition occurs in both cases at about 100°C (212°F). The rate of decomposition with temperature, however, is much higher for the nickel carbonyl than for the iron carbonyl; consequently, techniques had to be developed to control the mixtures of the two gases so that films of known composition could be deposited.

Handling metal carbonyls offers some difficulties. According to Badin et al,⁸ nickel carbonyl vapor explodes in air or oxygen at 20°C (68°F) and partial pressures as low as 15 mm Hg. Explosion is preceded by a variable induction period. Physiologically, nickel carbonyl is a toxic substance. The safety limit in air is 0.04 parts per million; hence it is essential to handle the carbonyls in a closed system with good ventilation.

4-3. DECOMPOSITION APPARATUS

To control, convey, and decompose the desired amounts of nickel and iron carbonyl vapor, several systems were developed. Apparatus I in figure 4-1 was used to plate in a vacuum. The nickel carbonyl was distilled in a high vacuum from a steel cylinder into a glass storage vessel which was cooled to 0°C (32°F). This technique both transferred the material out of the cylinder and purified it from any noncondensable gases. The carbonyl in the storage vessel I was transferred under a high vacuum through the

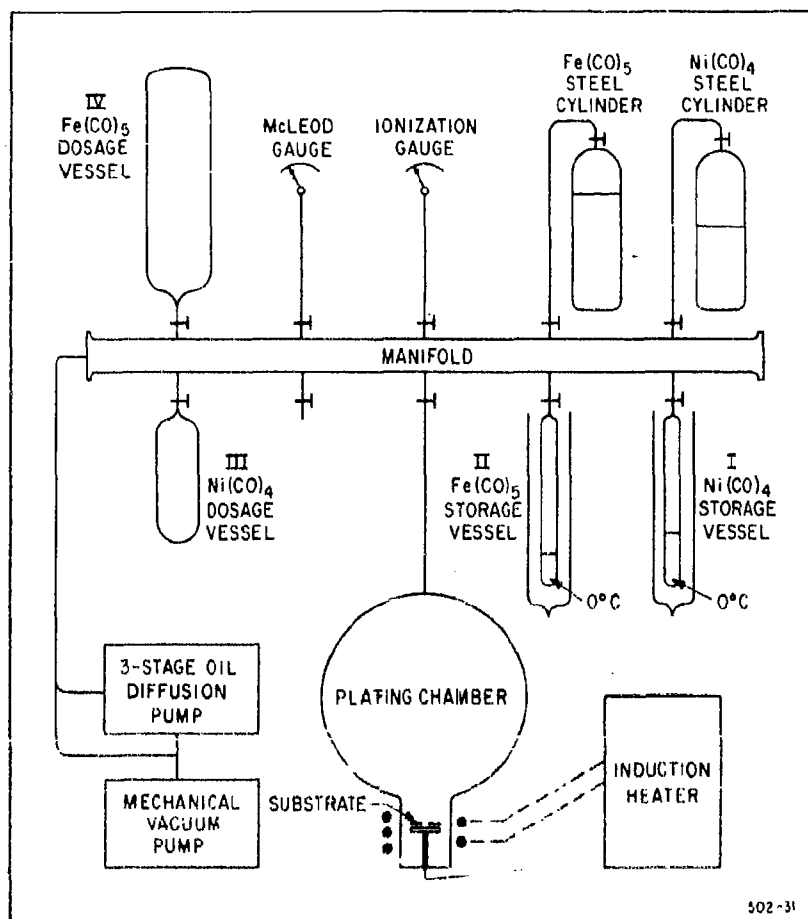


Figure 4-1. Apparatus I for Decomposing Nickel-Iron Carbonyl Mixtures

vacuum manifold into a pre-evacuated dosage vessel III. Similarly, a known quantity of the iron carbonyl was transferred from the steel cylinder into a storage vessel II and then into the dosage vessel IV. The carbonyls in the dosage vessels III and IV were then redistilled into the vacuum manifold and after allowing sufficient time to mix were sucked into the evacuated plating chamber. In the plating chamber a glass disc 9mm in diameter and 0.1 mm thick was placed on a metal platform and the metal platform was heated by induction to the decomposition temperature for the experiment. When the mixed carbonyl vapors came in contact with the heated glass substrate, a thin film of nickel-iron alloy was deposited.

Apparatus II, shown in figure 4-2, is a slight modification of apparatus I (figure 4-1). The prime modification is the introduction of an inlet for a carrier gas to transfer the mixed carbonyl vapors from the vacuum manifold to the plating chamber.

After several series of runs were made with the modified apparatus (II), it was found that further improvement in the equipment was necessary to improve the reproducibility of the films.

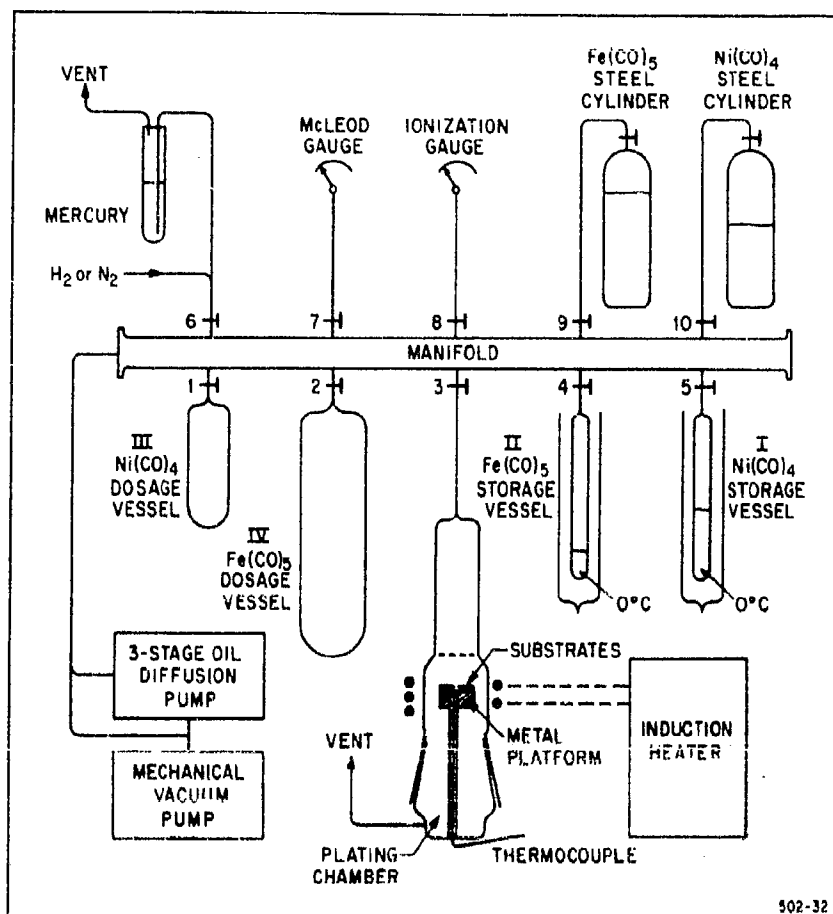


Figure 4-2. Apparatus II for Decomposing Nickel-Iron Carbonyl Mixtures

Two major changes therefore were introduced as shown in figures 4-3 and 4-4.

(1) The carbonyl mixture was prepared by saturating two streams of hydrogen with the individual carbonyl vapors. The temperature of the storage vessels containing the carbonyls was kept at fixed temperatures calculated from the vapor pressure equations to produce a known saturation of the hydrogen streams. The two streams then were mixed in a long tube connected to the plating chamber.

(2) The plating chamber itself was completely modified. A large brass disc 3 inches in diameter was mounted in a cylindrical chamber in such a way that it could be rotated by an externally revolving magnetic field. The disc contained 20 depressions in it so that twenty 9-mm cover-glass discs could be placed on it. The brass disc was heated by an induction heater. A thermocouple which was sealed into the chamber and which made sliding contact with the brass disc was used to monitor the temperature. Introducing these modifications resulted in a considerable improvement in the reproducibility of the films.

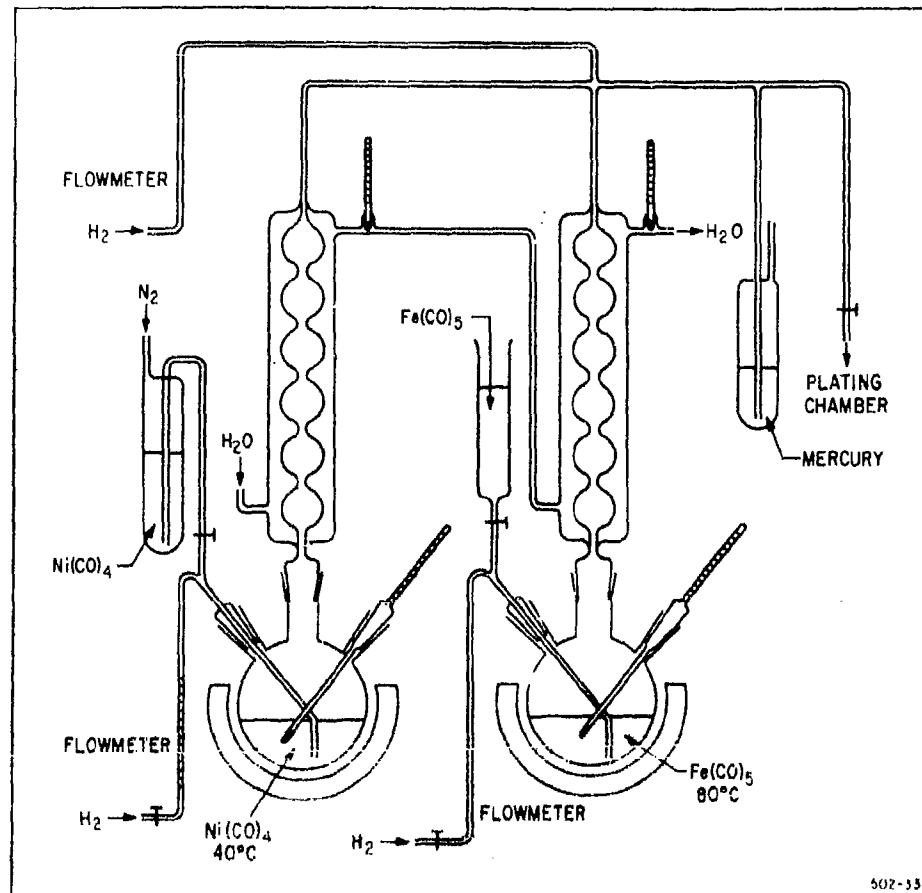


Figure 4-3. Apparatus for Preparing Nickel-Iron Carbonyl Mixtures

4.4. EFFECT OF DECOMPOSITION TEMPERATURE

A fixed mixture of nickel and iron carbonyls was introduced into the plating chamber and allowed to decompose onto the glass substrates held at temperatures of 300°C (572°F), 150°C (270°F), and 100°C (212°F). The results shown in table 4-1 indicate that there is a sharp dependence of film composition on the plating temperature. The percentage of iron in the film increases with increasing temperature. To obtain more information on this temperature dependence, the carbonyls were decomposed individually at several temperatures.

Discontinuous films of iron were found at 200°C (390°F), but continuous films were obtained at 300°C (572°F). Continuous films of nickel could be obtained at temperatures of 100°C (212°F). However, at 300°C filamentary growths of nickel were observed, indicating decomposition of the gas above the substrate.

Since the composition of the deposited film is both a function of the decomposition temperature and the composition of the carbonyl mixture, several runs were made with two different carbonyl mixtures over the temper-

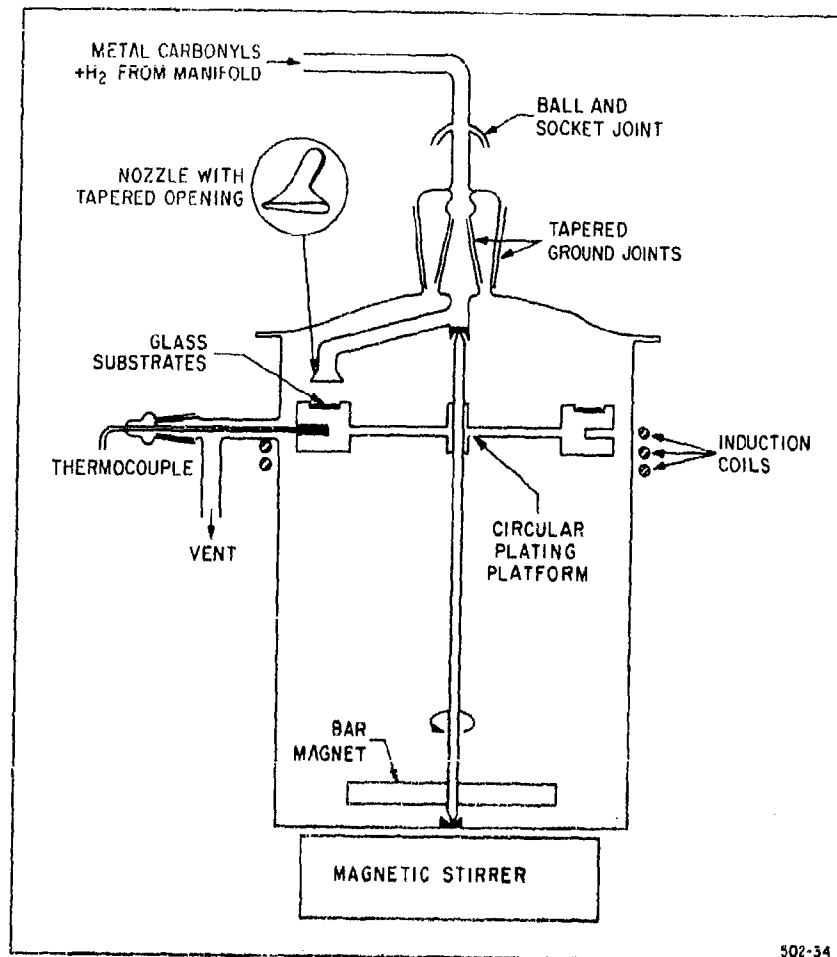


Figure 4-4. Rotating-Platform Apparatus for Decomposing Nickel-Iron Carbonyl Mixtures

ature range 100 to 300°C. Figure 4-5 indicates the dependence of composition of the films on variations in decomposition temperature and composition of the carbonyl mixture.

4-5. REPRODUCIBILITY OF FILM COMPOSITION

Figure 4-6 indicates the improvement in reproducibility of film composition as a result of the apparatus modifications mentioned in section 4-3. The standard deviation (σ) of the nickel content values of the films dropped from 11.4 to 1.7 as a result of using the rotating platform and the hydrogen streaming technique. In addition, the reproducibility of the magnetic properties of the films was improved greatly. In 19 of the 20 films in one run the coercivity varied ± 0.5 oersteds and in 15 films the variation was ± 0.25 oersteds. The thickness was 3340 to 3700 Ångstroms.

Table 4-1. Effect of Decomposition Temperature on Composition

Series	Decomposition Temperature		Nickel Content (percent)
	(°C)	(°F)	
180	300	572	26.8
181	300	572	26.5
161	200	392	42.9
162	200	392	43.0
170	150	302	65.1
171	150	302	66.1
172	150	302	79.3

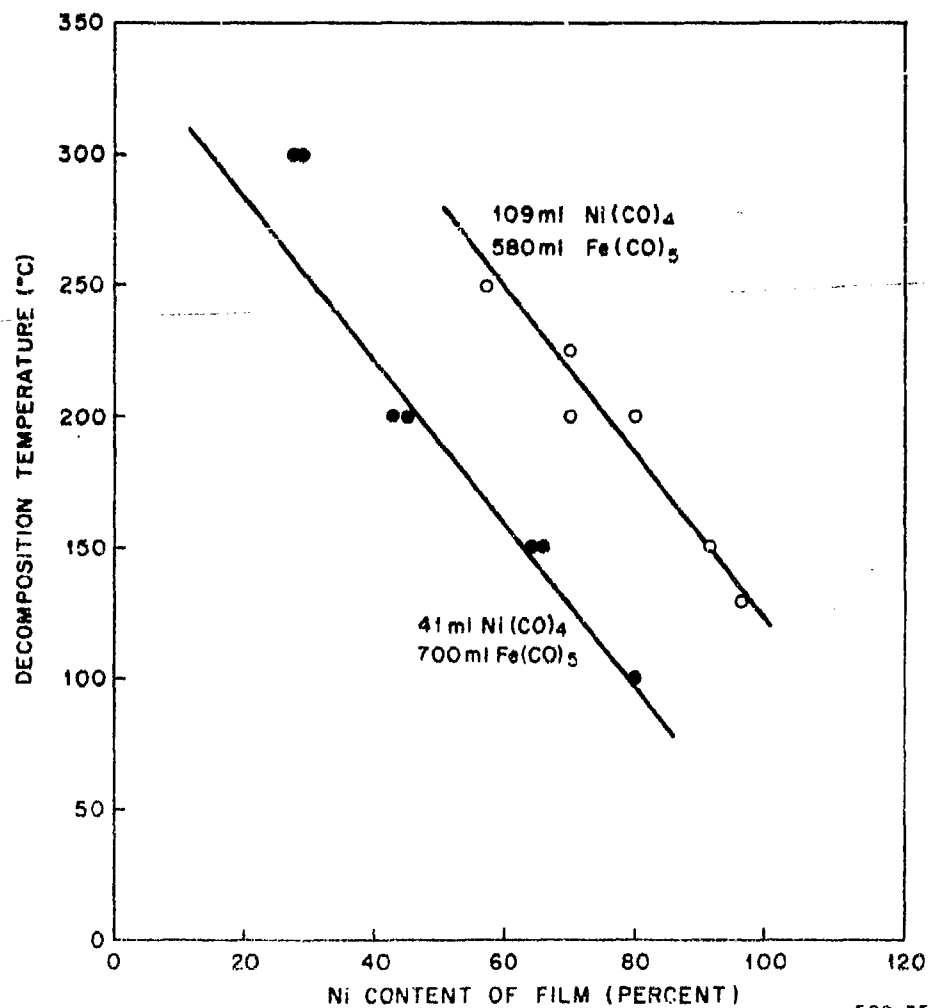


Figure 4-5. Effect of Decomposition on Nickel Content of Film

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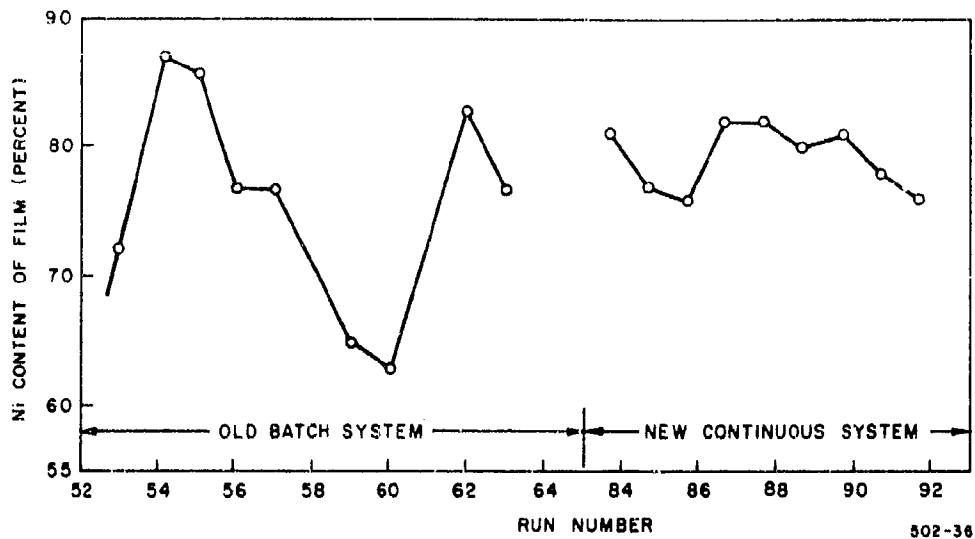
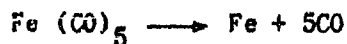
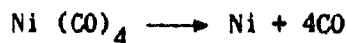


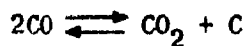
Figure 4-6. Variations in Nickel Content of Films in Two Different Systems

4.6. MAGNETIC PROPERTIES OF FILMS

Regardless of the temperature at which the carbonyl mixtures are decomposed the films usually have a high coercive force when tested. This high coercive force is attributed to the presence of carbon in the films. During the decomposition of the carbonyls according to the equation



the reversible reaction



also can take place.

The free carbon that is produced can go either into interstitial solution in the nickel-iron alloy or exist as discrete particles in the film, causing sufficient stress to raise the coercive force. Consequently, all of the films produced by the carbonyl process have to be annealed in order to lower the coercive force.

The annealing treatment consists of heating the films in a tube furnace in a hydrogen atmosphere for 30 minutes. The apparatus used for the annealing treatment is shown in figure 4-7. A rotating or linear magnetic field can be applied during the annealing treatment. If a rotating mag-

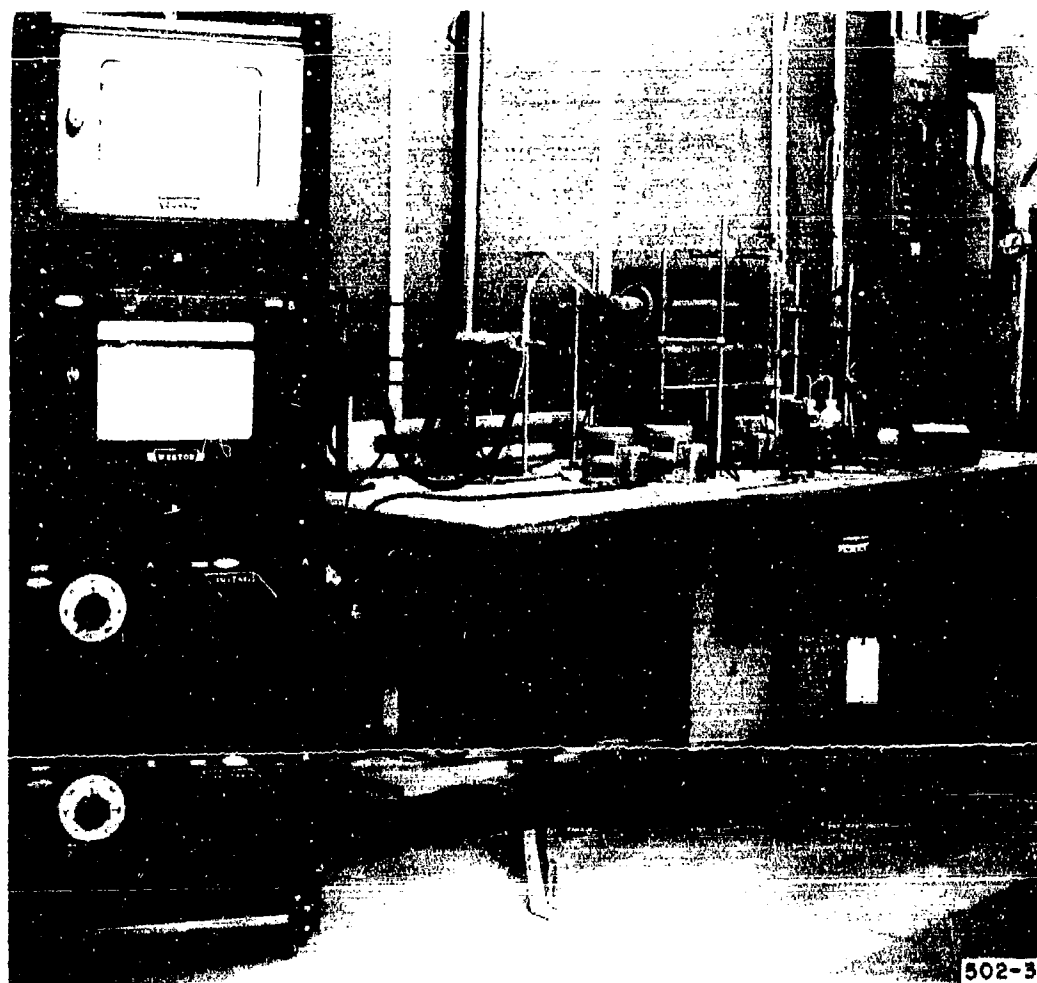


Figure 4-7. Apparatus for Annealing Carbonyl Films

netic field is applied, the films are magnetically isotropic as shown by the data in table 4-2. If the films are annealed in a linear magnetic field, a very definite anisotropy with a linear hysteresis loop in the hard direction is obtained as shown in figures 4-8a, b, and c. The hysteresis loop obtained by annealing in a rotating magnetic field is shown in figure 4-8d.

Annealing the films for 30 minutes in a hydrogen atmosphere at various temperatures in the range 50 to 550°C (122 to 1022°F) indicated a decrease in the coercive force at higher temperatures, as shown in figure 4-9.

Temperatures higher than 550°C could not be used because the glass substrate tends to soften and deform above 550°C. Annealing in a vacuum of 10^{-5} mm in the absence of an applied magnetic field at temperatures of 250, 400 and 550°C (472, 752, and 1022°F) lowers the coercive force in the easy direction and raises it in the hard direction as shown in figure 4-9. Above 400°C the heat-treated films show no anisotropy.

Table 4-2. Magnetic Properties of Carbonyl Films Annealed in Rotating and Linear Magnetic Fields

Annealing temperature: 550°C
 Annealing time: 30 minutes
 Annealing atmosphere: hydrogen

All data taken at 10-oersted drive

Series (CR-)	Thickness (Å)	Magnetic Properties			
		Easy Direction		Hard Direction	
		H _c (oe)	B _r /B _m	H _c (oe)	B _r /B _m

Rotating Magnetic Field

Annealing field strength: 36 oersteds

83/1	4110	2.4	0.96	2.4	0.96
84/1	3230	2.2	0.93	2.2	0.93
85/1	3390	2.7	0.93	2.7	0.93
151/1	3232	3.4	0.96	3.4	0.96
189/2	4750	2.0	0.92	2.0	0.92

Linear Magnetic Field

Annealing field strength: 300 oersteds

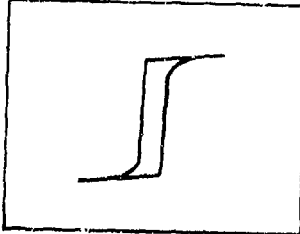
84/7	900	6.5	0.93	5.3	0.87
86/3	1300	4.5	0.98	2.2	0.96
26/6	1590	4.4	0.99	3.4	0.96
39/2	1720	4.0	0.97	2.1	0.91
92/3	2497	2.8	0.98	1.3	0.89
28/10	2900	2.8	0.95	0.9	0.67
19/6	3400	1.6	0.99	0.9	0.54

Annealing the films in a hydrogen atmosphere at a temperature of 550°C in a linear d-c field parallel to the plane of the film resulted in films that were anisotropic. The hysteresis loop data for films of this type are shown in table 4-2. The degree of anisotropy in these films is not clearly demonstrated by this data since the hysteresis loops in the hard directions begin to open up at relatively low drives. The anisotropy at low drives is evident in figure 4-8. Anisotropy produced by the magnetic anneal is much greater for films thicker than 2500 Ångstroms. Typical switching data for carbonyl films is shown in figure 4-10.

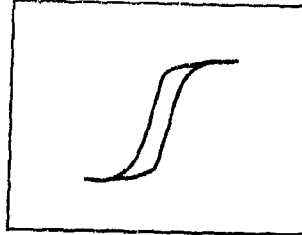
Certain films produced by the thermal decomposition of acetylacetonates and carbonyls possess unusual properties. In some of these films the easy and hard directions can be interchanged at room temperature by applying a magnetic field. Heretofore this had been accomplished only by the simultaneous application of heat and a magnetic field. These films have been called "threshold films".

SUBSTRATE : GLASS
ANNEALING TEMPERATURE : 550° C
ANNEALING ATMOSPHERE: HYDROGEN

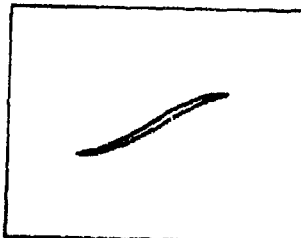
MAGNETIC FIELD : LINEAR



(a) Easy Direction: Drive = 10 Oe

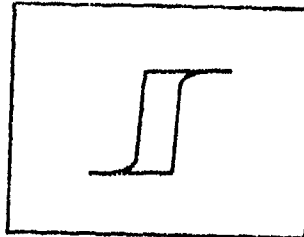


(b) Hard Direction: Drive = 10 Oe



(c) Hard Direction Drive = 2.5 Oe

MAGNETIC FIELD : ROTATING



(d) Drive = 10 Oe, No Anisotropy

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Figure 4-8. Hysteresis Loops, Carbonyl Films

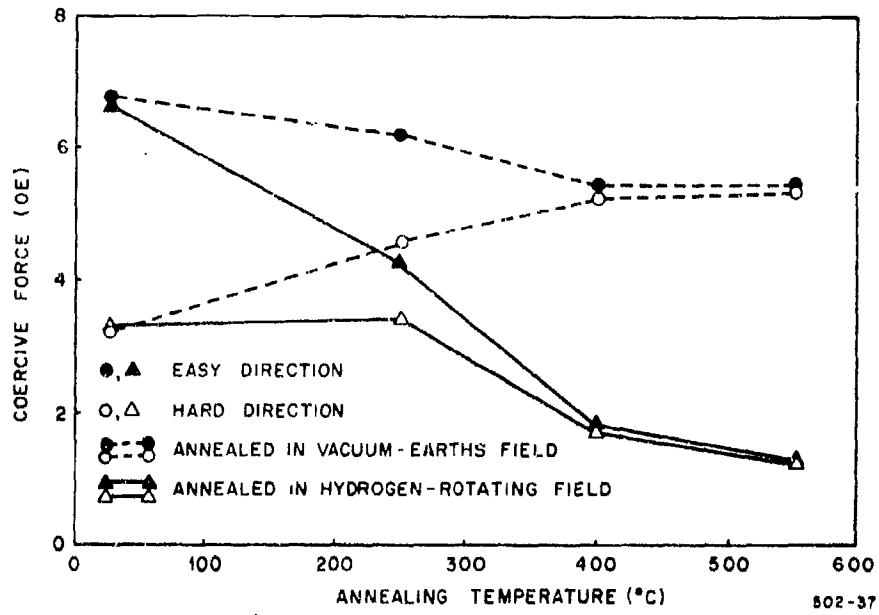


Figure 4-9. Effect of Annealing Temperature on Coercive Force

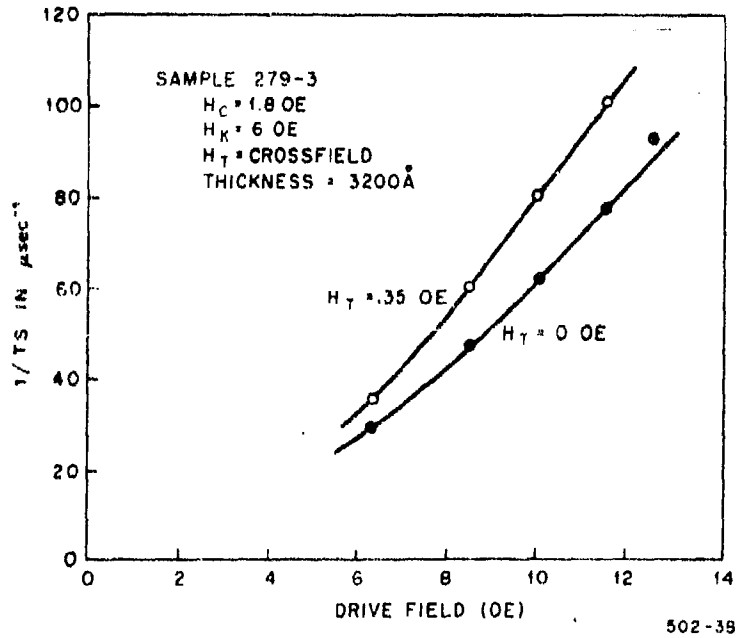


Figure 4-10. Switching-Time Curve for Carbonyl Films

4-7. CONCLUSIONS

As a result of this experimental work it has been demonstrated that nickel-iron films of low coercive force can be produced by the thermal decomposition of carbonyls. The conditions and the techniques for obtaining reproducible films have been determined. The magnetic properties of these films are not yet equal to those of electrodeposited and evaporated films, probably due to the presence of impurities. However, this process has shown much promise in the short time that it has been investigated and future research should be concentrated on the conditions necessary for obtaining high-purity films.

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