United States Patent

Pfeifer

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[54] NICKEL-IRON BASE MAGNETIC MATERIAL WITH HIGH INITIAL PERMEABILITY AT LOW TEMPERATURES

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[30] **Foreign Application Priority Data**

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[52] **U.S. Cl.** **148/31.55**, 75/170, 148/120, 148/121

[51] **Int. Cl.** **H01f 1/14**

[58] **Field of Search** 75/170; 148/31.55, 31.57, 100, 148/104, 120, 121

[56] **References Cited**

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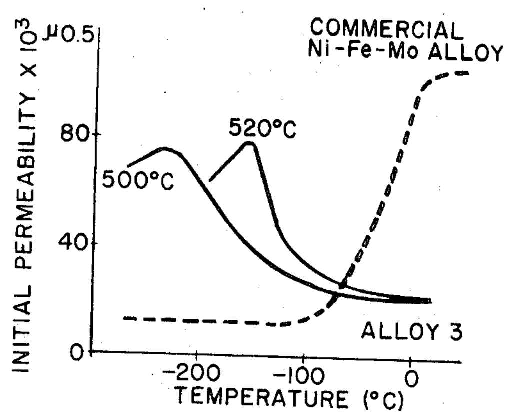
Attorney—F. Shapoe and R. T. Randig

[57] **ABSTRACT**

A nickel-iron base alloy is described to which controlled amounts of copper, manganese and molybdenum may be added. By proper selection of composition and heat treatment high initial permeabilities are obtained at cryogenic temperatures.

6 Claims, 9 Drawing Figures



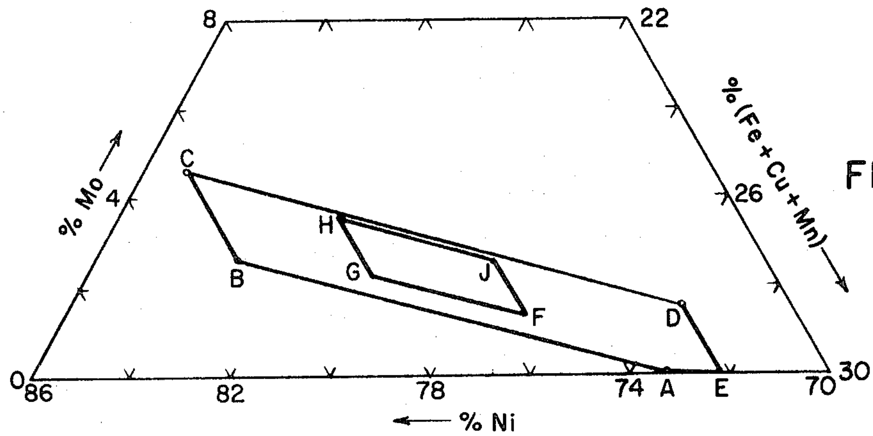


FIG. 1.

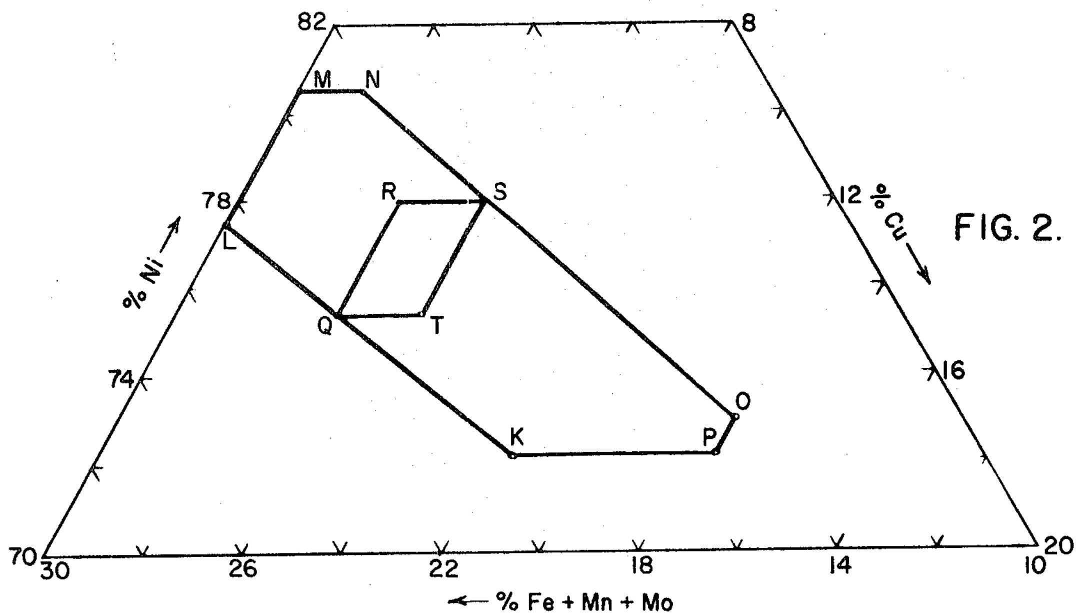


FIG. 2.

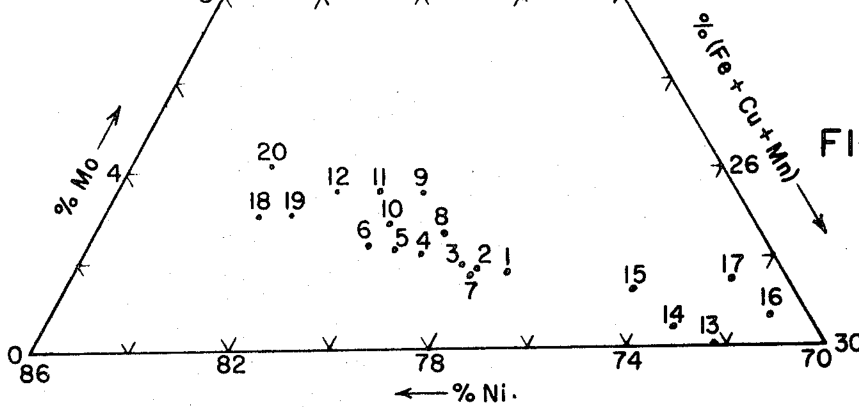


FIG. 3.

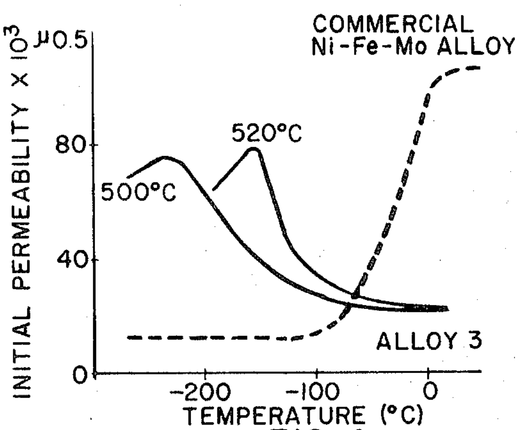


FIG. 4.

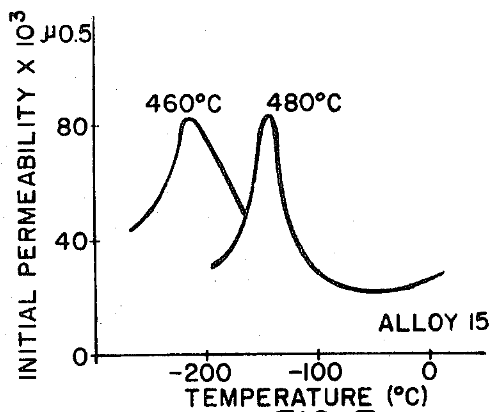


FIG. 7.

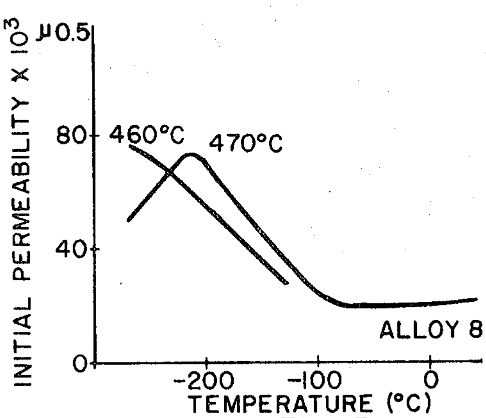


FIG. 5.

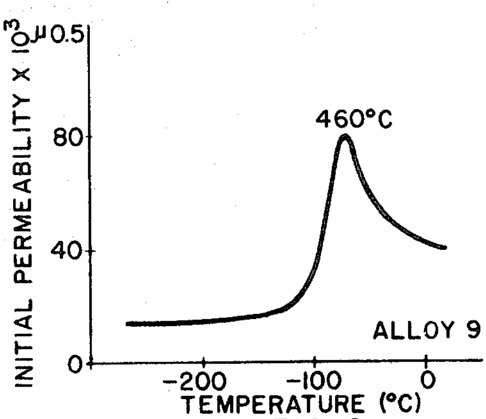


FIG. 8.

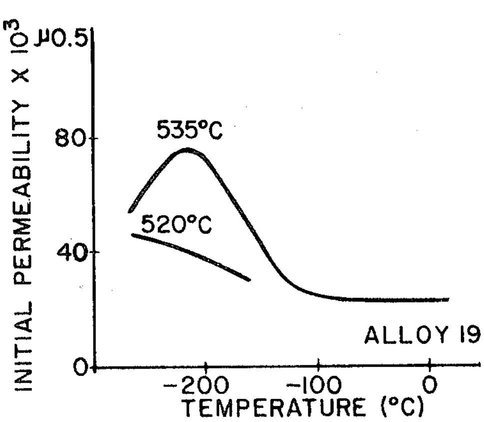


FIG. 6.

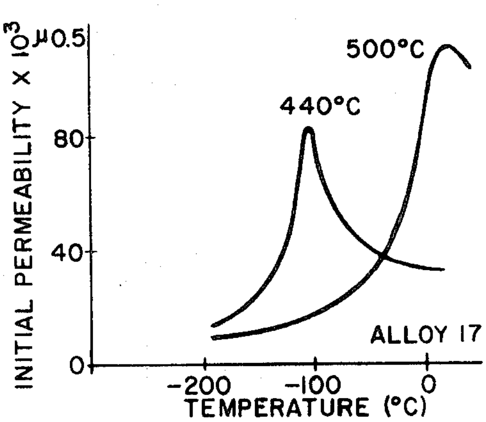


FIG. 9.

NICKEL-IRON BASE MAGNETIC MATERIAL WITH HIGH INITIAL PERMEABILITY AT LOW TEMPERATURES

BACKGROUND OF THE INVENTION

1. Field of the Invention

This invention relates to a magnetically soft material having a nickel-iron base which is characterized by a very high initial permeability at low temperatures, especially at temperatures below −180° C.

2. Description of the Prior Art

Suitable alloys with favorable magnetic properties at low temperatures are required in numerous fields of low temperature physics and technology. They are desirable, for example, for low temperature magnetic shields as well as for precision current instrument transformers and transmitters which operate at cryogenic temperatures such as obtained in liquid nitrogen or helium. For this type of application, the magnetically soft material must have as high as possible an initial permeability in the range of low temperatures. In magnetic shielding, the high initial permeability is a desirable condition for highly effective shielding against extremely weak extraneous magnetic fields. Moreover, for the current transformers, the indication or measuring error becomes smaller with increasing permeability of the transformer material, while in case of low current transmitters a very high inductivity can be obtained with a small number of turns if the transmitter material has a high permeability at small field intensities.

The high permeability magnetically soft materials which have been heretofore known have not met the above stated requirements. The relative permeability of the magnetically softest alloys with 70 to 80% of nickel when measured at room temperature and at a field intensity of 0.5 mOe (millioersted) is over 100,000 and decreases sharply with lower temperatures to about 10,000 to 15,000 at a temperature of −120° C.

In comparison, sendust alloys, that is, the ternary iron base alloys with about 7 to 14% of silicon and with about 2 to 7% of aluminum and which also fall into this category of the technology, reach higher permeability values even slightly below 0° C. but these higher permeability values are achieved in a relatively narrow range of temperature and these values are obtained only for field intensities of about 40 to 200 mOe, that is, for relatively high field intensities. Moreover, the point of maximum permeability depends specifically on the respective silicon and aluminum content of the alloy. For example, an iron-silicon-aluminum alloy with 9.9% of silicon and 5.6% of aluminum has at −100° C. a sharply defined maximum with a peak value of permeability of about 64,000 at a field intensity of 100 mOe. For field intensities below 30 mOe the permeability decreases to less than 15,000; moreover, for a field intensity of 1 mOe, the permeability amounts to even less than 10,000 (see "Journal of Physics," 1941, vol. IV, pages 569 to 572).

More importantly however the iron-silicon-aluminum alloys of the above specified composition are not suitable for many applications not only on the basis of their unfavorable dependency of their permeability on the field intensity but also because of their technological characteristics, that is, their high degree of brittleness (see C. Heck: "Magnetic Materials and Their Technical Applications," 1967, pages 403 and 404) does not permit any of the usual forming or cutting operations (other than grinding) and correspondingly there cannot be produced from these materials any strips, core laminations, or deep-drawn parts.

SUMMARY OF THE INVENTION

Therefore the purpose of this invention is to obtain in ductile alloys the best possible magnetically soft properties at low temperatures. The invention is based specifically on the problem of preparing a nickel-iron base magnetic material which has, at temperatures below −180° C., a relative initial permeability of more than 40,000 in weak magnetic fields.

Broadly speaking this is accomplished by selecting the alloying components such that there is present between about 8.9% and 27.6% iron, up to 12.5% copper, up to 4.6% molybdenum, from about 0.2% to about 1.0% manganese and the balance essentially nickel with incidental impurities. The alloy is thereafter processed by hot and cold working and the finish gauge material is thereafter annealed at a temperature within the range between about 1,050° and about 1250° C. for a time period of between about 2 hours, and, about 8 hours followed by cooling to room temperature. The annealed material is then subjected to a final heat treatment at a temperature between the Curie temperature and 550° C. for a time period of between about 1 hour and about 5 hours followed by quenching to room temperature.

An object of the present invention is to provide a ductile nickel-iron base alloy having high initial permeability at cryogenic temperatures and at low field intensities.

A specific object of the present invention is to provide a ductile nickel-iron base magnetic material and a heat treatment therefor whereby the material will exhibit an initial permeability of at least 40,000 in a field intensity of 0.5 mOe at a temperature of less than −180° C.

Other objects of the present invention will become apparent to those skilled in the art when read in conjunction with the following description and the drawings in which:

BRIEF DESCRIPTION OF DRAWINGS

FIG. 1 is a ternary diagram illustrating the broad and preferred limits of the alloying components;

FIG. 2 is a ternary diagram illustrating the relation of the copper content with respect to the balance of the alloying components;

FIG. 3 is similar to FIG. 1 but illustrating the actual composition of alloys made and tested as set forth in Table I; and

FIGS. 4 through 9 inclusive illustrate the relationship between initial permeability and temperature for alloys 3, 8, 19, 15, 9 and 17 respectively of Table I.

DESCRIPTION OF PREFERRED EMBODIMENT

The alloy of the present invention is a nickel-iron base alloy to which controlled amounts of at least one of copper, manganese and molybdenum are added. While in broad general terms the alloy contains, in percent by weight, from about 8.9% to about 27.6% iron, up to 12.5% copper, up to 4.6% molybdenum, from about 0.2% to about 1.0% manganese and the balance nickel incidental impurities, the alloying components must be balanced in accordance with the circumscribed areas of FIGS. 1 and 2.

More particularly, according to this invention the actual composition of the magnetic material lies within or in immediate vicinity of a range of multicomponent system nickel-(iron + copper + manganese)-molybdenum which is defined in FIG. 1 by the polygon

A (73.3% Ni; 26.7% (Fe+Cu+Mn); 0% Mo)-
B (80.5% Ni; 16.9% (Fe+Cu+Mn); 2.6% Mo)
C (80.5% Ni; 14.9% (Fe+Cu+Mn); 4.6% Mo)
D (72.2% Ni; 26.3% (Fe+Cu+Mn); 1.5% Mo)
E (72.2% Ni; 27.8% (Fe+Cu+Mn); 0% Mo)-A.

The preferred range of alloying components is defined by the polygon

F (75.4% Ni; 23.2% (Fe+Cu+Mn); 1.4% Mo)
G (78.0% Ni; 19.7% (Fe+Cu+Mn); 2.3% Mo)
H (78.0% Ni; 18.5% (Fe+Cu+Mn); 3.5% Mo)
J (75.4% Ni; 22.1% (Fe+Cu+Mn); 2.5% Mo)-F

with the restriction that the content of manganese is 0.2 to 1.0% in each instance. Moreover, the content of copper associated with the content of nickel must be within the range which in FIG. 2 is determined by the polygon

K (72.2% Ni; 19.4% (Fe+Mn+Mo); 8.4% Cu)
L (77.5% Ni; 22.5% (Fe+Mn+Mo); 0% Cu)
M (80.5% Ni; 19.5% (Fe+Mn+Mo); 0% Cu)
N (80.5% Ni; 18.1% (Fe+Mn+Mo); 1.4% Cu)
O (73.0% Ni; 14.5% (Fe+Mn+Mo); 12.5% Cu)

P (72.2% Ni; 15.3% (Fe+Mn+Mo); 12.5% Cu)-K.

Preferred results are obtained where the copper content is maintained within the polygon

Q (75.4% Ni; 21.3% (Fe+Mn+Mo); 3.3% Cu)
R (78.0% Ni; 18.7% (Fe+Mn+Mo); 3.3% Cu)
S (78.0% Ni; 17.0% (Fe+Mn+Mo); 5.0% Cu)
T (75.4% Ni; 19.6% (Fe+Mn+Mo); 5.0% Cu)-Q of FIG. 2.

When the alloying components are balanced within the circumscribed areas of FIGS. 1 and 2, the material is annealed during its manufacture in a non-oxidizing atmosphere for several hours, specifically 2 to 8 hours, at 1,050° to 1,250° C. and afterwards it is subjected to final heat treatment for several hours, specifically 1 to 5 hours, at a temperature between the Curie temperature and 550° C.

Improvement of the magnetic properties in the range of low temperatures obtained by the selection of alloying and heat treatment can be seen from the following examples. Reference is directed to Table I and to FIG. 3 which tabulates the chemical composition of a number of alloys which were made and tested.

TABLE I
[Chemical composition (percent by weight)]

Alloy No.	Ni	Fe	Cu	Mo	Mn	Si
1	75.55	18.00	4.20	1.65	0.63	Trace
2	76.05	17.25	4.37	1.75	0.60	0.02
3	76.30	16.70	4.48	1.86	0.66	0.01
4	77.05	16.08	4.45	2.05	0.69	0.01
5	77.50	15.30	4.50	2.23	0.61	Trace
6	77.95	14.80	4.40	2.32	0.63	0.02
7	76.35	16.50	4.75	1.60	0.63	0.01
8	76.30	15.35	4.80	2.60	0.63	0
9	76.25	15.30	4.40	3.50	0.48	Trace
10	77.30	14.95	4.40	2.80	0.77	0
11	77.10	14.40	4.35	3.50	0.48	0.01
12	78.00	13.30	4.45	3.50	0.48	Trace
13	72.20	14.95	12.30	0	0.77	0
14	72.80	15.15	10.75	0.41	0.78	0
15	73.20	14.35	10.70	1.25	0.78	0
16	70.75	14.10	13.60	0.72	0.82	0.02
17	71.05	13.40	13.60	1.50	0.72	0.05
18	79.80	16.50	~0.10	3.00	0.77	0
19	79.15	17.15	0	3.05	0.75	Trace
20	79.05	16.25	0	4.1	0.70	Trace

In a vacuum furnace there were produced 20 nickel-iron alloys with the chemical composition given in per cent by weight in Table I and indicated by the same numbers in the nickel-(iron + copper + manganese)-molybdenum alloying diagram in FIG. 3. After forging, the ingots were hot rolled to a thickness of 2.5 mm followed by annealing at 1,050° C. Thereafter, the material was cold rolled to a final thickness of 0.1 mm with intermediate annealing where necessary. From the strip 0.1 mm thick and 10 mm wide there were produced wound ring cores of 25 mm inside diameter and 35 mm outside diameter. These cores were then annealed for 5 hours at 1,200° C. in pure hydrogen, cooled to room temperature, subjected to final heat treatment in hydrogen at the time and temperatures set forth in Table II and subsequently the cores were quickly brought to room temperature.

The permeability ($\mu_{0.5}$) of the ring cores was determined by a Maxwell bridge at field intensity of 0.5 mOe and at the frequency of 70 Hz. These conditions closely approximate the initial permeability. For measuring the dependency of the permeability of the temperatures in the range between −268.9° C. and +20° C., the cores were placed in a suitable protective copper casing, cooled in a cryostat to the temperature of liquid helium (i.e. −268.9° C.) or liquid nitrogen (i.e. −195.8° C.) and then permitted to warm naturally while the permeability and temperatures were monitored. The measurements of the temperatures below −200° C. were made by means of a goldiron-chromel thermocouple and above −200° C. by means of a copper-constantan thermocouple which was placed in the protective casing.

The test results reproduced in Table II and the permeability values shown in FIGS. 4 to 7 as a function of temperature demonstrate that the magnetic alloys with nickel-iron base obtained according to this invention and when heat treated as set forth herein have in the range of low temperatures a far higher initial permeability than the presently available high grade commercial nickel-iron materials whose maximum permeability is in the range of room temperature as it is shown by the dotted curve in FIG. 4.

TABLE II

Alloy No.	Final heat treatment: Time (hrs.)	Final heat treatment: Temperature (° C.)	Maximum initial permeability at, 0.5 m0e.	Test temperature (° C.)	Permeability 0.5 m0e. at— −269° C.	−196° C.	−100° C.	Shape of permeability, temperature curve
1	2	520	68,000	−120	----------	30,000	53,000	
	2	500	54,000	−269	54,000	44,000	29,000	Maximum below −269° C.
2	2	520	76,000	−125	----------	30,000	52,000	Narrow maximum.
	2	500	70,000	−230	60,000	60,000	32,000	Broad maximum.
3	2	520	78,000	−155	----------	63,000	34,000	(Fig. 4).
	2	500	75,000	−230	68,000	60,000	28,000	Do.
4	2	550	70,000	−165	26,000	44,000	26,000	Narrow maximum.
	2	520	74,000	−210	48,000	69,000	32,000	Broad maximum.
5	2	550	66,000	−185	38,000	64,000	32,000	Do.
	2	520	85,000	−240	75,000	60,000	27,000	Do.
6	2	550	68,000	−269	68,000	46,000	25,000	Maximum below −269° C.
	2	520	----------	----------	----------	27,000	17,000	
7	2	520	87,000	−180	40,000	80,000	43,000	Broad maximum.
	2	510	88,000	−255	87,000	68,000	24,000	Do.
8	2	470	74,000	−215	50,000	68,000	25,000	(Fig. 5).
	2	460	76,000	−269	76,000	53,000	----------	Do.
9	2	460	79,000	−70	15,000	15,000	32,000	(Fig. 8).
10	2	490	109,000	−205	46,000	100,000	27,000	
	2	480	99,000	−245	68,000	52,000	18,000	
11	2	460	92,000	−140	14,000	16,000	19,000	Very narrow maximum.
12	2	480	81,000	−165	----------	52,000	32,000	
	2	460	72,000	−269	72,000	38,000	20,000	Maximum under −269° C.
13	2	520	61,000	−175	23,000	48,000	----------	
	2	500	60,000	−215	28,000	54,000	17,000	
14	2	500	69,000	−195	36,000	69,000	20,000	
	2	480	----------	----------	----------	32,000	14,000	
15	2	480	83,000	−140	----------	30,000	28,000	(Fig. 7).
	2	460	82,000	−215	42,000	70,000	----------	Do.
16	2	550	80,000	−15	----------	12,000	19,000	
	2	440	87,000	−155	----------	43,000	36,000	Very narrow maximum.
17	2	500	110,000	+20	----------	8,000	17,000	(Fig. 9).
	2	440	82,000	−105	----------	14,000	80,000	Do.
18	2	550	49,000	−269	49,000	43,000	22,000	Maximum under −269° C.
19	2	535	76,000	−215	53,000	69,000	14,000	(Fig. 6).
	2	520	46,000	−269	46,000	36,000	----------	Do.
20	2	480	69,000	−240	62,000	59,000	32,000	Broad maximum.

From the test data in Table II and from the permeability-temperature curves in FIGS. 4 to 9 it can be seen how the position and the form of the maxima of permeability depend on the composition of the alloys and on the heat treatments. As a principle for selection of a suitable material with easy magnetization at low temperatures it was found that in general the maximum of permeability is shifted toward the low temperatures by a lower final heat treatment temperature and also by longer annealing times. To avoid the Perminvar effect the final heat treatment must be at a temperature above the Curie temperature.

Another relationship found to exist in the alloy of the present invention is that where the molybdenum content increases toward the upper limit at any given nickel level the final heat treatment temperature must decrease toward the Curie temperature in order to achieve the high initial permeability. Also, where the alloy has a composition near the line F–G of FIG. 1 increasing nickel contents require higher final heat treatment temperatures approaching 550° C. in order to obtain the high initial permeability at low temperatures.

The maximum initial permeability of the alloys found outside of the range limit A – B – C – D – E – A (Nos. 9, 11, 16 and 17) cannot be shifted into the temperature range of liquid nitrogen and of liquid helium by a final heat treatment at a lower temperature.

The advantage obtained with the invention consists in making available a more ductile magnetically soft material which has a very high permeability at the low temperatures, especially in the range between −180° and −269° C. The relationships which have been found permit the selection of the alloy and the final heat treatment so that the highest possible permeability or a permeability of predetermined value in a given range may be selected at will.

The magnetic materials according to this invention with the nickel-iron base with high permeability at low temperatures are suitable above all for the low temperature cooled magnetic shields, current transformers, and transmitters as well as for relays, magnetic switches, memories, and multipliers.

I claim as my invention:

1. A heat treated ductile nickel-iron base magnetic alloy consisting essentially of, by weight, from about 8.9% to about 27.6% iron, up to about 12.5% copper, up to about 4.6% molybdenum, from about 0.2% to about 1.0% manganese and the balance essentially nickel, the alloy exhibiting maximum initial permeability at subzero temperatures when the alloying components within the ranges set forth hereinbefore are balanced to provide an alloy having a composition within the area ABCDEA of FIG. 1 and in which the copper content is balanced with respect to the remainder of the alloying components to provide an alloy having the composition falling within the area KQLMNSOPK of FIG. 2, and the alloy has been given a final heat treatment at a temperature within the range between the Curie temperature and 550° C.

2. The alloy of claim 1 in which the copper content is balanced with respect to the remainder of the alloying component to provide an alloy having the composition within the area QRSTQ of FIG. 2.

3. The alloy of claim 1 in which the alloying components are balanced to provide an alloy having the composition within the area FGHJF of FIG. 1.

4. A readily workable heat treated nickel-iron base alloy containing copper, molybdenum and manganese, the alloying components being selected to provide an alloy having a composition falling within the area FGHJF of FIG. 1, the copper content is selected with respect to the remaining elements to provide a composition within the area QRSTQ of FIG. 2 and the alloy has been given a final heat treatment at a temperature within the range between the Curie temperature and 550° C, said alloy being characterized by exhibiting its maximum initial permeability at a temperature of below about −100° C.

5. A magnetic core suitable for use at temperatures below about −100° C. having a high initial permeability and formed from the alloy of claim 1.

6. A heat treated ductile nickel-iron base magnetic alloy consisting essentially of, by weight, from 8.9 to 27.6% iron, up to 12.5% copper, up to 9.6% molybdenum, from 0.2% to 1.0% manganese and the balance nickel with incidental impurities, the alloy exhibiting maximum initial permeability at a temperature below −100° C. when the alloying components within the ranges set forth hereinbefore are balanced to provide an alloy having a composition within the area FGHJF of FIG. 1 and in which the copper content is balanced with respect to the remainder of the alloying components to provide an alloy having the composition falling within the area KQLMNSOPK of FIG. 2 and the alloy has been given a final heat treatment at a temperature within the range between about 440° C. and about 550° C.

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