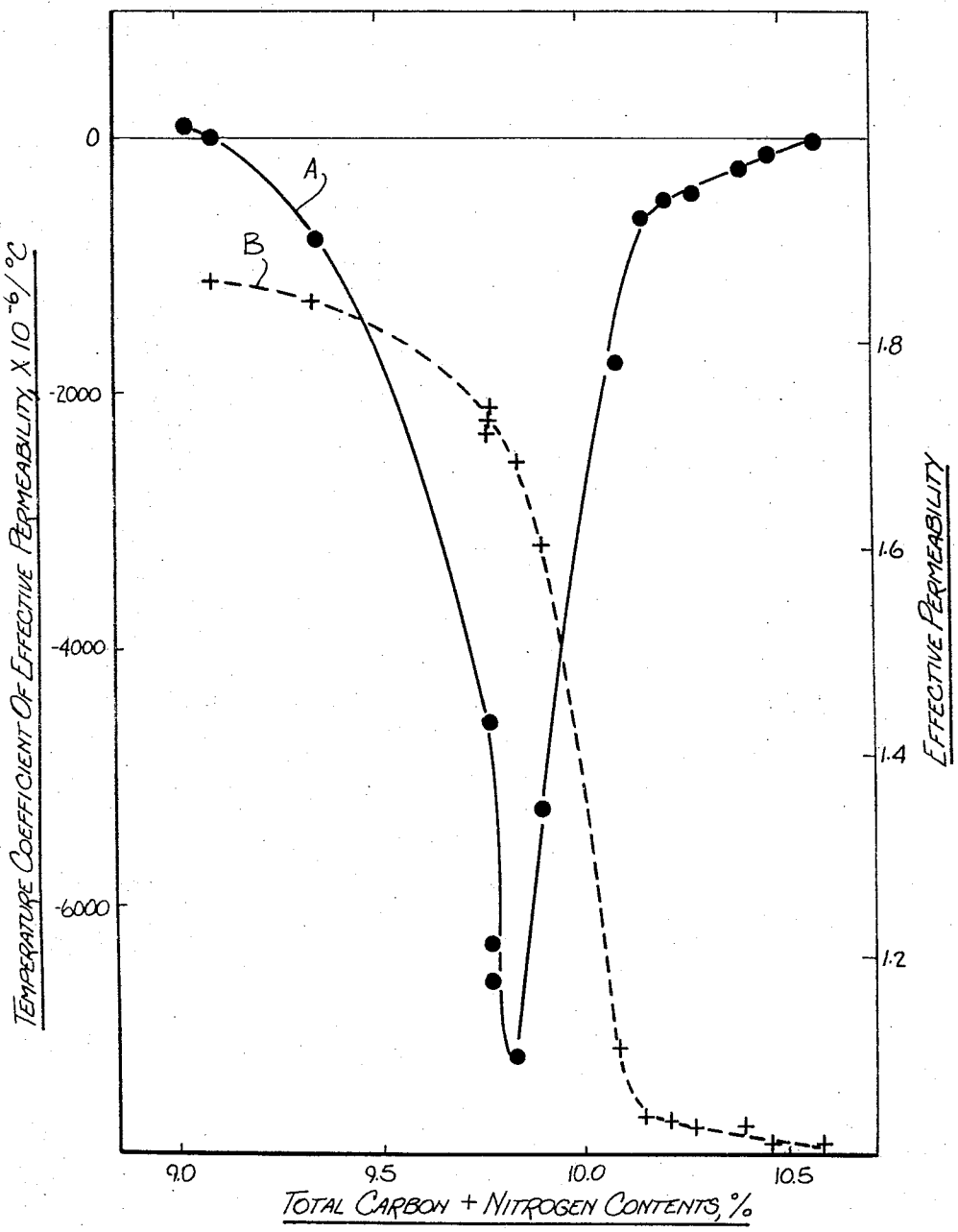
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IRON POWDER AND CORE WITH CONTROLLED PERMEABILITY COEFFICIENT

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IRON POWDER AND CORE WITH CONTROLLED PERMEABILITY COEFFICIENT

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The present invention relates to production of iron power inductance cores and more particularly to production of nitrogen-containing carbonyl-iron powder and of inductance cores comprising such powder, which powder and cores are characterized by controlled temperature coefficients of permeability.

It is well known that carbonyl-iron powder, that is to say, powder produced by the thermal decomposition of iron carbonyl, is commonly used as the material of magnetic cores for electrical purposes. Carbonyl-iron powder usually contains small amounts of both carbon and nitrogen. At the present time, grades of carbonyl-iron powder commercially available have carbon and nitrogen contents as follows:

Percent C:	Percent N
<0.05	<0.02
0.65	0.65
0.9	0.9
1.4	1.8
2.2	2.6

In all these grades the oxygen content is less than 0.5%.

In the manufacture of magnetic cores, carbonyl-iron powder particles, which are of extremely small particle size, are normally first coated with a thin film of insulation, a binder is then added, and finally the powder is compressed into a solid mass. When made of commercially available carbonyl-iron powder, such as the aforedescribed powder, the resultant core has a high Q value and a positive temperature coefficient of permeability, e.g., $20\times10^{-6}/°$ C.

In many frequency-selective circuits incorporating an inductor, it is desirable that the temperature coefficient of an inductor comprising a coil and a core be near zero, or be negative so as to compensate for the positive value of the temperature coefficient of other components, and thus provide that the frequency characteristics of the circuit remain constant irrespective of temperature fluctuation over some predetermined temperature range. Difficulties are encountered in attempting to provide inductors which are characterized by negative temperature coefficients of permeability and thus are capable of compensating for positive temperature coefficients of permeability in other portions of a circuit. Accordingly, where inductor cores having temperature coefficients of permeability of small positive values, or zero, or negative values are needed, the high positive temperature coefficients of permeability of commercially available carbonyl-iron powders are serious disadvantages of these powders.

Although many attempts were made to overcome the foregoing difficulties and other difficulties and disadvantages, none, as far as I am aware, was entirely successful when carried into practice commercially on an industrial scale.

It has now been discovered that in an inductive electric circuit a low temperature coefficient of permeability which is only a little greater than zero, or zero, or lower

can be achieved with an inductor core which comprises special carbonyl-iron powder.

It is an object of the present invention to provide an inductor core characterized by a low temperature coefficient of permeability.

Another object of the invention is to provide a process of making an inductor core characterized by a low temperature coefficient of permeability.

The invention also contemplates providing a special carbonyl-iron powder core which is characterized by a temperature coefficient of permeability of zero or less than zero.

It is a further object of the invention to provide a special nitriding process for producing nitrided carbonyl-iron powder which contains a special controlled amount of carbon.

The invention further contemplates providing a new process whereby a body comprising special carbonyl-iron powder is used as an inductor core.

Other objects and advantages will become apparent from the following description taken in conjunction with the accompanying drawing which depicts the correlation of magnetic permeability and temperature coefficient of permeability characteristics to the total nitrogen plus carbon content of carbonyl-iron powder in accordance with the invention.

Generally speaking, the present invention contemplates production of novel inductor cores characterized by low temperature coefficients of permeability which are not greater than about $5\times10^{-6}/°$ C., i.e., temperature coefficients of permeability which are of negative values or zero or of small positive values up to only about plus $5\times10^{-6}/°$ C. As referred to hereinafter, magnetic permeability is the effective permeability referred to a particular shape of core when magnetically coupled to a particular coil and temperature coefficient of permeability is the average temperature coefficient of said magnetic permeability over the temperature range about 20° C. to about 75° C. Cores of the invention comprise special carbonyl-iron powder, which special powder has a total nitrogen plus carbon content of at least 9.0% and not greater than about 10.5%.

A surprising discovery of the invention is that carbonyl-iron powder with contains nitrogen shows most striking variations in the temperature coefficient of permeability which depend on the combined nitrogen and carbon content. The foregoing is illustrated in the drawing, wherein Curve A depicts the relationship of temperature coefficient of permeability to total nitrogen plus carbon content of inductor cores of carbonyl-iron powder in accordance with the invention. It should be pointed out that temperature coefficient of permeability is essentially independent of frequency up to frequencies of at least 10 megacycles per second. Curve A demonstrates that the compositional range of about 9.0% to about 10.5% total nitrogen plus carbon content for carbonyl-iron powder of the invention is a critical range of composition and that cores of carbonyl-iron powder within said range are characterized by a temperature coefficient of permeability not greater than zero. The curve is especially steep in the range 9.6% to 10% total nitrogen plus carbon and in accordance with the invention this range is a special advantageous critical range of composition for carbonyl-iron powder characterized by a low negative temperature coefficient of permeability not greater than about minus $1600\times10^{-6}/°$ C. and as low or lower than minus $7000\times10^{-6}/°$ C., e.g., minus $7200\times10^{-6}/°$ C. The microstructure of the

powder is that associated with one wholly of epsilon phase.

The present invention also contemplates a new process for preparing nitrogen-containing powder of the invention by nitriding carbonyl-iron powder in a special rapidly flowing ammonia atmosphere whereby carbonyl-iron powder which as a core is characterized by a low temperature coefficient of permeability is produced by nitriding carbonyl-iron powder to a predetermined extent.

In producing carbonyl-iron powder having a nitrogen plus carbon content of 9.0% to 10.5% by the process of the invention, it is advantageous to use as the starting material one of the carbonyl-iron powders containing specific amounts of carbon and nitrogen and not those substantially free from carbon and nitrogen, since the latter (containing less than 0.05% carbon and less than 0.02% nitrogen) are more dfficult to nitride. For achieving best results in commercial operation to produce inductor cores having controlled temperature coefficients of permeability not greater than about $5\times10^{-6}/°$ C., the present invention provides a new nitrogen-containing carbonyl-iron powder containing about 7% to about 10% nitrogen, about 0.5% to about 2.0% carbon and having a total nitrogen plus carbon content of 9.0% to about 10.5%.

The special carbonyl-iron powder of the invention can be produced by nitriding commercially available carbonyl-iron powders. The nitriding can be effected by heating the powder in ammonia. In accordance with the nitriding process of the invention, carbonyl-iron powder that initially contains at least about 0.02% nitrogen and at least about 0.05% carbon is heated in a furnace or reactor at about 400° C. to about 500° C. for about 4 hours to about 20 hours in a rapidly flowing atmosphere of ammonia, which rapidly flowing atmosphere flows at a rate of not less than 25 liters per hour per square inch of cross-sectional area of the furnace measured perpendicular to the direction of flow. After the powder is nitrided, it is advantageously cooled rapidly while in the ammonia atmosphere in order to avoid decomposition of the powder, which is not particularly stable and is disposed to lose nitrogen more readily than to gain it at the nitriding temperature. In a typical process, carbonyl-iron powder containing 0.65% carbon and 0.65% nitrogen is placed in a boat with a copper bottom and this boat is then placed in a furnace which has previously been flushed with nitrogen to remove all oxygen. The powder is then heated in this furnace in an atmosphere of ammonia at 450° C. for a period of 4 to 6 hours. The ammonia is caused to flow rapidly through the furnace and the direction of the flow is reversed halfway through the heating. If the ammonia is not driven rapidly through the furnace, then there is a risk of prior partial decomposition of the ammonia accompanied by the concentration of hydrogen rising with consequential partial removal of nitrogen and carbon from the powder. It is important to maintain the powder in the atmosphere of ammonia until it is cool. One way of doing this is to provide a water-cooled zone as an extension of the furnace and, when enough nitrogen has been introduced into the powder, to pull the boat to this zone where its copper bottom is effective to remove heat rapidly from the powder. The size of an average particle of powder of the invention is from about 2 microns to about 7 microns.

According to the invention, carbonyl-iron powder is nitrided to a combined nitrogen and carbon content of 9% to 10.5%. There is a peak in the negative coefficient at 9.8%, the coefficient rising steeply to this peak in the range from 9.6% to 10.0%, as illustrated in the drawing.

By mixing the novel nitrided powder in appropriate amounts (in accordance with the combined nitrogen and carbon content) with unnitrided carbonyl-iron powder, it is possible to produce a core having zero or a predetermined negative coefficient.

The present invention provides a special agglomerate body especially adapted for use as an inductor core having a low and controlled temperature coefficient of permeability. Agglomerate bodies of the invention comprise particulate matter and electrically insulating material. At least a portion of the particulate matter of a body of the invention is special carbonyl-iron powder having a total nitrogen plus carbon content of 9.0% to 10.5%. Electrically insulating material is disposed as a coating surrounding the carbonyl-iron powder particles of the body and a binding ingredient can also be included in the body. The materials of the body are mixed and pressed together to form an agglomerate mass for the special body. Agglomerate bodies of the invention, which bodies are characterized by temperature coefficients of permeability not greater than about $5\times10^{-6}/°$ C., are either magnetic or nonmagnetic depending upon the total nitrogen plus carbon content.

For the purpose of giving those skilled in the art a better understanding of the invention and/or a better appreciation of the advantages of the invention, the following illustrative examples are given:

Example I

About 70 grams of carbonyl-iron powder containing about 0.8% nitrogen and about 1.0% carbon and of an average particle size of about 3.8 microns was placed in a boat three inches long by two inches wide forming a bed one-half inch deep and this boat was then charged into a tube furnace having an average cross-sectional area of about four square inches, which furnace had been previously flushed with nitrogen to remove all oxygen. Ammonia was introduced into the furnace at a flow rate of about 200 liters per hour. The carbonyl-iron powder was nitrided by heating at a temperature of about 450° C. in the ammonia atmosphere for about five hours while maintaining the aforesaid flow rate of ammonia. Thereafter the powder was rapidly cooled in the ammonia atmosphere to about room temperature. The nitrided carbonyl-iron powder produced by this exemplary process in accordance with the invention had a nitrogen content of about 8.95% and a carbon content of about 0.83%. A magnetic inductor core of this nitrided powder was produced by coating particles of this powder with insulating films of sodium silicate (1% by weight), bonding with phenol formaldehyde resin (3% by weight) and pressing the coated particles at a pressure of 25 long tons per square inch to agglomerate them into a body having a cylindrical shape with a diameter of ⅜ inch and a length of ½ inch. The insulating film was applied by techniques well known to those skilled in this art. The pressed core was heat treated for two hours at 130° C. to polymerize the bonding resin. The magnetic permeability of this core was 1.72 and the temperature coefficient of permeability of the core was minus $6500\times10^{-6}/°$ C.

Example II

Nitrided carbonyl-iron powder of Example I was used as an additive in the production of a magnetic core characterized by a preselected temperature coefficient of permeability not greater than zero by adding about 0.7 gram of nitrided powder of Example I to about 100 grams of unnitrided carbonyl-iron powder containing about 0.75% nitrogen and about 0.68% carbon and mixing these two powders. An inductor core in accordance with the invention was made by coating particles of the mixture with insulating films of sodium silicate (1% by weight), bonding with phenol formaldehyde resin (3% by weight) and pressing the particles into a bonded agglomerate body having cylindrical shape with a diameter of ⅜ inch and a length of ½ inch. The magnetic permeability of the core was about 2.12 and the temperature coefficient of permeability was about minus $0.6\times10^{-6}/°$ C.

Further examples of nitrogen-containing carbonyl-iron powder mixtures for inductor cores in accordance with the invention are set forth hereinafter in Table I:

TABLE I

Mix.	High Nitrogen plus Carbon Powder			Low Nitrogen plus Carbon Powder		
	Grams	Percent N	Percent C	Grams	Percent N	Percent C
A	5.2	8.49	0.89	100	0.75	0.68
B	0.7	9.07	0.73	100	0.75	0.68
C	14	9.50	0.79	100	0.75	0.68
D	67	9.50	0.79	100	0.75	0.68

As referred to herein, "high nitrogen plus carbon powder" is carbonyl-iron powder containing a total of at least 9.0% nitrogen plus carbon and "low nitrogen plus carbon powder" is carbonyl-iron powder containing less than 9.0% nitrogen plus carbon. All percentage compositions set forth herein are percentages by weight.

Characteristics of inductor cores comprising powder mixtures of Table I are set forth in Table II:

TABLE II

Mix.	Permeability	Temperature Coefficient of Permeability, $\times 10^{-6}$/°C.
A	2.1	−6.5
B	2.1	0
C	2.0	−0.4
D	1.7	−104

Although the insulating and bonding materials in the foregoing examples of inductor cores in accordance with the invention are sodium silicate and phenol formaldehyde resin, respectively, it is to be understood that the insulation which is disposed on carbonyl-iron particles and the binder which gives adhesion to the insulated particles in the agglomerate bodies for inductor cores of the invention can be of other such materials which are known to those skilled in the art of producing carbonyl-iron powder inductor cores, e.g., phosphoric acid may be used as the insulator and an epoxy resin as the binder. In producing an inductor core having a preselected and controlled temperature coefficient of permeability in accordance with the invention by using high nitrogen carbonyl-iron powder as an additive for controlling said coefficient, the proportions of high nitrogen powder and low nitrogen powder are selected by emperical methods.

Magnetic permeability of cores made from the nitrided iron powders of the invention is related to the total nitrogen plus carbon content of the carbonyl-iron powder of the said cores. Curve B of the drawing is typical of the relationship of the magnetic permeability of cores of powder of the invention. Following Curve B from left to right, it is evident that permeability decreases with increasing nitrogen plus carbon content and that the magnetic properties are continually diminishing until the core ceases to be magnetic and the permeability is unity. Weakly magnetic and even nonmagnetic carbonyl-iron powder of the invention is of use in the manufacture of inductor cores for operation at very high frequency.

The Q values of the special nitrogen-containing powders are low compared with those of a normal carbonyl-iron powder; however, in order to produce a core of zero temperature coefficient of permeability the nitrided powders need only be added in small amounts to the base powder. Consequently the Q values of the base powder are little effected.

The present invention is particularly applicable to production of carbonyl-iron powder and powder mixtures and of inductor cores comprising carbonyl-iron powder for use in frequency-selective electrical circuits such as inter alia professional discriminatory electric circuits which are subject to temperature fluctations with change of environment and yet need to operate satisfactorily at all times.

In view of the foregoing it is apparent that the present invention provides a method for producing an electromagnetic inductance that does not increase with increases in temperature of the inductor materials, or increases at only a small rate not greater than about 5×10^{-6}/° C., from about 20° C. to about 75° C. by using an agglomerate body comprising a special carbonyl-iron powder which has an average total nitrogen plus carbon content of about 9.0% to about 10.5% as an inductor core. A body of the invention can be used as an inductor core by coupling the body with the electromagnetic field produced by passing an electric current through an electrically conductive element which is disposed in close proximity to the body in accordance with methods known to those skilled in the art of producing inductances by using materials of the prior art for inductor cores.

Although the present invention has been described in conjunction with preferred embodiments, it is to be understood that modifications and variations may be resorted to without departing from the spirit and scope of the invention, as those skilled in the art will readily understand. Such modifications and variations are considered to be within the purview and scope of the invention and appended claims.

I claim:

1. A process of using as an inductor core an agglomerate body containing iron powder comprising inductively coupling an agglomerate powder body containing carbonyl-iron powder consisting essentially of 7% to about 10% nitrogen, about 0.5% to about 2% carbon, with a total nitrogen plus carbon content of 9.0% to about 10.5%, and the balance essentially iron and having a microstructure consisting essentially of epsilon phase with an electrically conductive element disposed in close proximity to said body by passing an electric current through said electrically conductive element to characterize said core by a temperature coefficient of permeability not greater than 5 times 10^{-6} per ° C.

2. A process as set forth in claim **1** wherein the total nitrogen-plus-carbon content of the powder is 9.6% to 10%.

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