Patented Jan. 19, 1954

2,666,724

UNITED STATES PATENT OFFICE

2,666,724

PROCESS OF PREPARING IRON POWDER OF IMPROVED ELECTROMAGNETIC PROPERTIES

Hans Beller, Cranford, N. J., assignor to General Aniline & Film Corporation, New York, N. Y., a corporation of Delaware

No Drawing. Application December 3, 1952, Serial No. 323,943

3 Claims. (Cl. 148—16.6)

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This invention relates to iron powder of improved electromagnetic properties and particularly to the process of preparing the same.

It is known that electromagnetic cores prepared from powdered carbonyl iron can be utilized in high frequency coils, transformers, antenna loops, and the like. The purpose of such cores is to assure a high Q value or to provide an inductance control or both. The finely divided iron employed in the manufacture of the electromagnetic cores is frequently obtained by the thermal decomposition of iron pentacarbonyl as described in United States Patent 1,759,659. The iron powder obtained according to the process described in the patent is usually in the form of microscopic spheres each of which is composed of several hundred submicroscopic crystals. These contain 1 to 2% of impurities, i. e., approximately 0.2% of oxygen, 0.7% of carbon, and 0.5% of nitrogen. The powder is prepared by introducing the iron carbonyl into a heated vessel in such a manner that the decomposition takes place substantially in the free space of the vessel instead of by contact with the heated walls of the vessel.

Cores made from such a powder will impart to coils with which they are magnetically coupled a certain additional inductance and also a certain additional series resistance. If the additional inductance due to the core be called L′ and the additional series resistance due to the core be called R′, the final inductance L″ and the final series resistance R″ of the coil-core assembly will be:

$$L''=L+L'$$
$$R''=R+R'$$

wherein L and R refer to the inductance and resistance of the coil without core. If the coil is operated at a frequency f, one can use the so-called Q-value, i. e., the ratio of inductive reactance to series resistance, which thus becomes for the coil:

$$Q=\frac{2\pi fL}{R}$$

and for the coil-core assembly:

$$Q''=\frac{2\pi fL''}{R''}=\frac{2\pi f(L+L')}{R+R'}$$

In a great many applications the designer or engineer tries to obtain (1) the maximum possible L″ with (2) the minimum possible R″. Conditions (1) and (2) may be combined in postulating simply the maximum possible Q″.

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By processing a given powder with the required care and know-how, relatively high Q″ values can be obtained. But each given powder has a definite upper limit of Q values for each frequency and coil above which no presently known method can raise it.

It is also known that reactions involving iron and nitrogen have been studied and are discussed in standard handbooks, such as J. W. Mellor's "A Comprehensive Treatise on Inorganic and Theoretical Chemistry," vol. VIII (March 1947 edition), pp 131–136 and Gmelin's Handbuch der Anorganischen Chemie, 8 te Auflage, Eisen, by R. J. Meyer et al., 1936, (A) p. 1718–1729, (B) p. 137–156.

The action of ammonia on iron was first observed in 1808. Powdered iron for nitriding experiments was first used in 1865. Powdered carbonyl iron (reduced or desoxidized was first mentioned, in connection with ammonia treatments, by A. Mittasch et al. in Z. Elektrochemie 34, 833 (1928). The existence of bulk Fe_4N and its crystal structure was first described by L. Bonnerot, Comptes Rendus 158, 996 (1914) and substantiated by G. Hagg, Nature 121, 826 (1928). The combination of ammonia and hydrogen was first mentioned by C. B. Sawyer in Trans. Am. Inst. Min. Met. Engin. 69, 808 (1928). Nitrides containing a certain amount of carbon were described by A. Fry in Stahl and Eisen, 43, 1273 (1923). The after-treatment technique in nitrogen, in order to homogenize the crystals is mentioned by H. Braune iin Jernkontorets Ann. p. 656 (1906). The ferromagnetic nature of nitrides derived from reduced carbonyl iron powder was proved by E. Lehrer in Zs. Techn. Physik 10, 184 (1929). The increase of resistivity of iron with increasing nitrogen content has also been observed. A factor of 3.23% per 0.01% is given.

Since these earlier works, there have been a number of magnetic and physicochemical studies of nitrides, some of which again used reduced carbonyl iron powders, for example, V. G. Paranjpe et al., Transactions AIME, 188, 261 (1950); A Chretien et M. Mathis, Comptes Rendus 228, 91 (1949); C. Guillaud et H. Creveaux, Comptes Rendus 222, 1170 (1946); and K. H. Jack, Proc. Roy. Soc. A195, 34 (1948).

None of the previous works discloses high frequency properties or suggests that reduced, crude, carbonyl iron powders can be processed into a much improved high frequency material by a heat treatment at temperatures ranging from 450° to 500° C. in an atmosphere of ammonia gas to yield a powder having increased Q

values when employed as cores of high frequency coils.

It is an object of the present invention to provide an improved process of preparing ferromagnetic powder having increased Q values above their hitherto attainable limits.

Other objects and advantages will appear from the following description.

The ferromagnetic powder having increased Q values is prepared from any non-reduced carbonyl iron powder obtained by the thermal decomposition of iron pentacarbonyl by subjecting it to a heat treatment in a closed furnace at a temperature ranging from 450° to 500° C. in an atmosphere of ammonia gas for a period of time ranging from 4 to 10 hours. Thereafter, the iron powder is cooled in a stream of dry nitrogen gas and utilized as such or subjected to a milling treatment.

In practicing the process of the present invention, iron powder obtained by the thermal decomposition of iron pentacarbonyl, as described in United States Patent 1,759,659, is placed in boats or trays which are heated in a muffle furnace in an atmosphere of ammonia gas. The gas is led through the muffle and the temperature maintained between 450° to 500° C., preferably at 500° C., for a period of time ranging from 4 to 10 hours. After this period, the muffle furnace is allowed to cool to about room temperature and the ammonia gas displaced by dry nitrogen. The cooling usually takes from 6 to 12 hours during which the dry nitrogen is passed through the muffle. The powder is then removed from the muffle furnace and milled, if desired, in a ball mill in order to break up clusters that may have formed by sintering until a powder composed substantially of discreet spheres of practically the original size results. It is to be noted that the ball-milling treatment is only employed if the treated powder shows slight sintering.

The rate of flow of ammonia gas over the trays containing the powder is maintained at a total flow of 0.5 to 2 cubic feet per minute of the gas per 100 lbs. of powder used. The pressure of the gas is approximately atmospheric. The ammonia gas treatment is completed within 4 to 10 hours usually between 6 to 8 hours.

The magnetic permeability of the powder obtained in accordance with the foregoing process, is essentially the same as that of the untreated powder and, hence, ranges from 6.0 to 13.0.

The following example will describe in detail the methods for accomplishing the above objects, but it is to be understood that it is inserted merely for the purpose of illustration and is not to be construed as limiting the scope of the invention.

EXAMPLE

A tray containing 250 grams of iron powder obtained by the thermal decomposition of iron pentacarbonyl, was placed in a muffle furnace. The furnace was heated to a temperature of 500° C. for 7 hours, while .0035 cubic feet/minute (100 cc./min.) of ammonia gas passed over the powder. The furnace was cooled, while permitting a stream of dry nitrogen gas to pass through the muffle, until approximate room temperature was

attained. The cooling took about 8 hours. The slightly sintered powder was removed from the furnace and milled with 1 kilogram of ¾" steel balls for 1 hour at 75 R. P. M. Finally it was passed through a 180-mesh screen. No coarse particles were detected.

For the electromagnetic measurements, 180 grams of the treated powder, and the same sample weight of the original powder, i. e., untreated, were separately placed in individual test tubes which formed the removable cores of a set of coils. The following table illustrates the increased Q value of the treated powder over that of the untreated powder.

Q (relative values)

	5 mc.	25 mc.
Untreated	99	101
Treated	115	146

The above values were determined in a Boonton Q-meter from which Q values and tuning capacitances are read directly.

The meaning of the term "Q value" as used herein has been defined heretofore by a formula. From the formula it can be readily seen that the value of "Q" will generally increase with an increase of coil inductance and decrease with an increase in coil resistance. The addition of an iron core will increase the inductance but also introduce eddy current and other losses. These losses act as and can be represented as an increase in effective coil resistance.

From the foregoing table, it is clearly evident that the process of the present invention results in the improvement of Q values at 5 mc. by 16% and at 25 mc. by 45%. These increased Q values were not previously obtainable with currently available iron powders.

While I have disclosed the preferred embodiments of my invention, it will be readily apparent to those skilled in the art that many changes and variations may be made therein without departing from the spirit thereof. Accordingly, the scope of the invention is to be limited solely by the appended claims.

I claim:

1. The process of improving the electromagnetic properties of carbonyl iron powders which comprises heating pulverulent iron obtained by the thermal decomposition of iron pentacarbonyl at a temperature ranging from 450° to 500° C. in the presence of ammonia gas for a period of time ranging from 4 to 10 hours, and cooling the heated powder with dry nitrogen.

2. The process according to claim 1, wherein the heating is conducted at 500° C.

3. The process according to claim 1, wherein the cooling with dry nitrogen is followed by a ball milling treatment.

HANS BELLER.

OTHER REFERENCES

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