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TREATMENT OF FINELY DIVIDED MAGNETIC MATERIAL

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FIG. 1

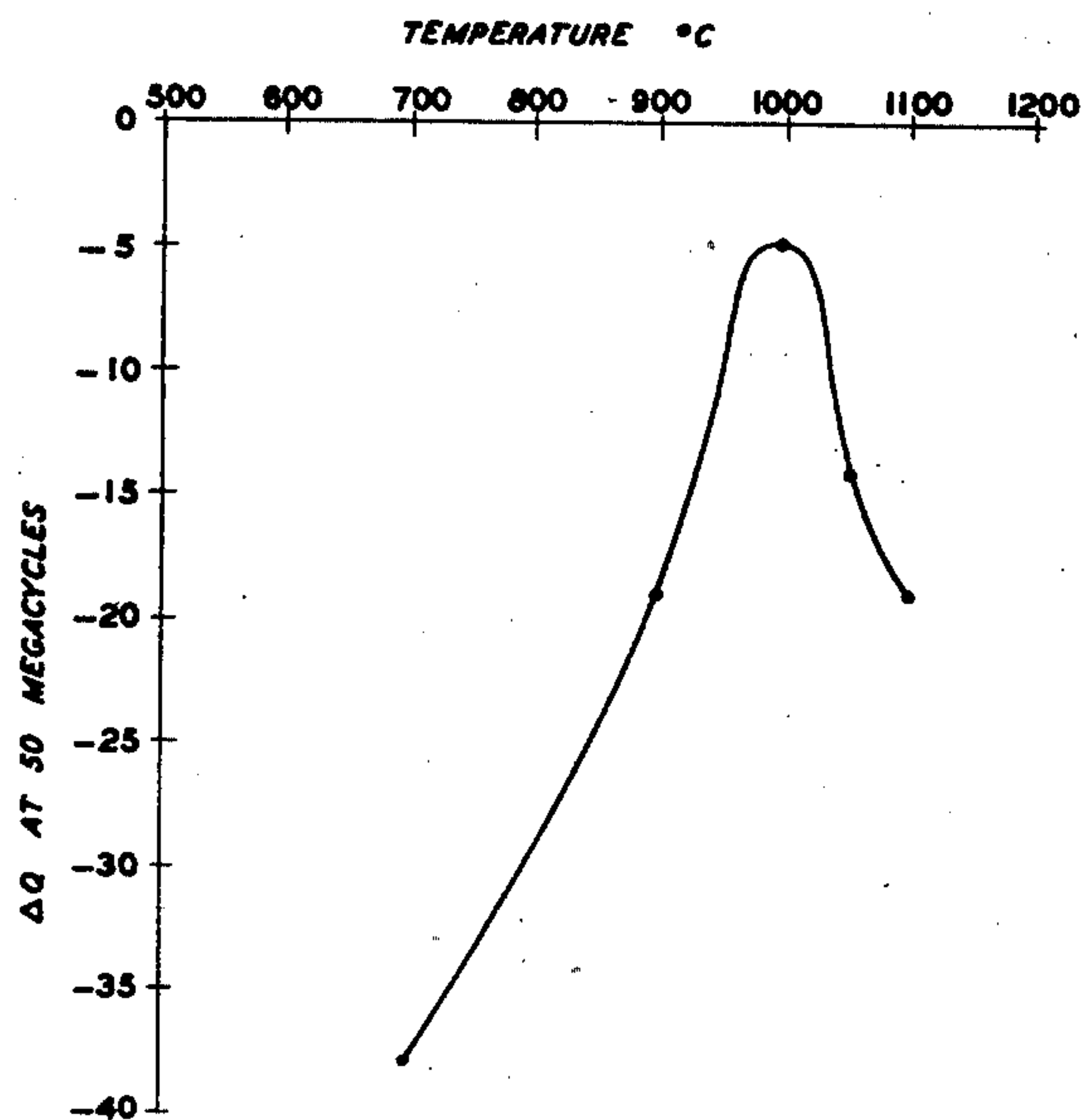
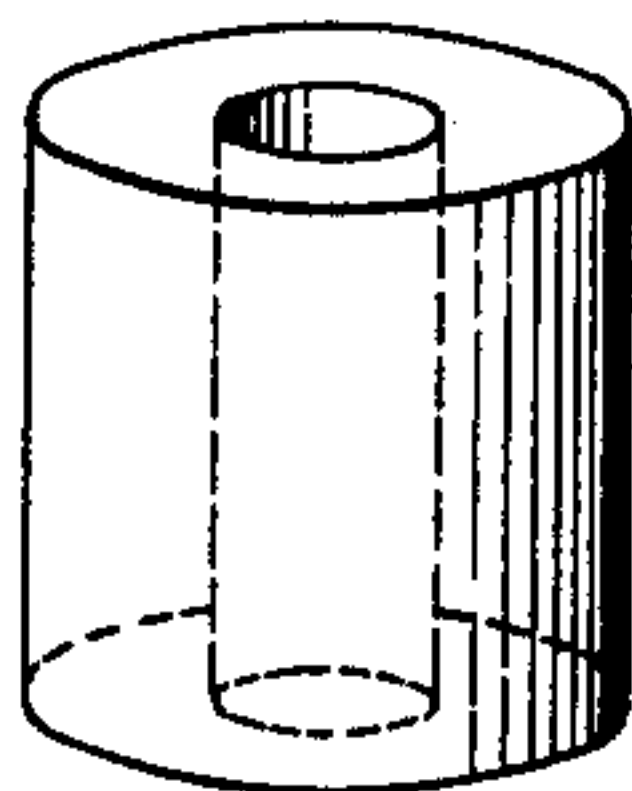


FIG. 2



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TREATMENT OF FINELY DIVIDED MAGNETIC MATERIAL

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This invention relates to magnetic bodies comprising finely divided magnetic material, and more particularly to the treatment of finely divided magnetic material to improve its properties for use in such magnetic bodies.

Finely divided magnetic material to the treatment of which the present invention pertains may be employed in magnetic bodies of the compressed type, i. e., those formed by compressing a finely divided magnetic material, with or without an added electrical insulating material, under high pressures to form a body of dense structure. Said finely divided magnetic material may also be employed in magnetic bodies made of loosely assembled particles of magnetic material with or without added electrical insulating material therebetween. The magnetic bodies formed of the finely divided magnetic material treated according to the present invention may be employed to particularly great advantage in high frequency electrical circuits operating at frequencies of at least about 25 megacycles; they may also be employed, although to less advantage, in circuits operating at lower frequencies such as telephone, broadcast radio and other circuits.

Because of the characteristics of the magnetic material treated according to the present invention, it may be employed to particular advantage in compressed magnetic cores for inductance coils in high frequency circuits. For the purpose of illustration but not of limitation the magnetic material treated according to this invention will be largely discussed hereinafter in connection with such cores.

It has been proposed to employ an oxide of the ferro-ferric oxide type in magnetic bodies. However, it has been found that such an oxide, even if synthetically produced and hence presumably produced under controlled conditions which will provide uniformity of composition and substantial freedom from impurities, does not provide as good results as are desired when the oxide is employed in magnetic cores for inductance coils, particularly those cores employed in high frequency circuits. In cores employed in such circuits losses in the cores must be low; this and other magnetic characteristics of the cores should be highly uniform among all cores of the same type which are manufactured; and the characteristics of the cores should not change substantially during operation. It has been found that cores formed of the commercially available finely divided ferroso-ferric oxides often have loss characteristics which are excessively high; in other cases cores formed of such oxides which have initially satisfactory low loss characteristics develop excessive losses and deleterious changes in other magnetic characteristics during operation, particularly when the cores are em-

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ployed under conditions at which they reach relatively high temperatures and even more particularly when employed under conditions of varying temperatures; in other cases cores formed of ferroso-ferric oxides have losses which are initially excessively high and become progressively worse during operation.

In accordance with the present invention, these disadvantages of magnetic bodies comprising finely divided ferroso-ferric oxide are minimized, if not entirely eliminated, by the employment of a ferroso-ferric oxide which has been treated according to the present invention. According to the present invention, finely divided ferroso-ferric oxide of a particular composition indicated hereafter is heated in an inert atmosphere to an elevated temperature which is advantageously in a critical temperature range. Such heat treatment of the oxide reduces excessive losses which might be present in cores formed of the untreated oxide, and stabilizes the magnetic characteristics of the cores so that their losses do not increase and their other magnetic characteristics do not deleteriously change during operation even at elevated or varying temperatures.

The exact nature and advantages of the present invention will be better understood from the following detailed description in connection with the accompanying drawings in which:

Fig. 1 represents a curve illustrating the reduction in losses which may be obtained in magnetic cores formed of finely divided ferroso-ferric oxide treated according to the present invention; and

Fig. 2 illustrates one form of core formed of finely divided ferroso-ferric oxide treated according to the present invention.

Ferroso-ferric oxide theoretically has a composition corresponding to the chemical formula Fe_3O_4 and is usually considered to be a ferrous ferrite of the formula $\text{FeO} \cdot \text{Fe}_2\text{O}_3$. In the process of the present invention, however, the ferroso-ferric oxide heat treated is one in which the Fe_2O_3 component is in substantial molecular excess with respect to the FeO component; that is, the molal ratio of $\text{Fe}_2\text{O}_3/\text{FeO}$ is substantially greater than one. As an example, marked advantages are provided if, in a ferroso-ferric oxide heat treated according to the present invention, the molal ratio $\text{Fe}_2\text{O}_3/\text{FeO}$ lies between about 1.25 and 1.75; exceptionally good results have been obtained with finely divided ferroso-ferric oxides having a molal ratio of between about 1.40 and 1.55. Moreover, the ferroso-ferric oxide is substantially free of impurities; it advantageously contains less than about 0.03 per cent by weight of metallic iron and only comparable amounts of other impurities when it is to be employed in cores used in high frequency cir-

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cults. Higher proportions of impurities might impair magnetic characteristics at high frequencies of about 25 megacycles and up. The size of the particles of the finely divided ferroso-ferric oxide does not appear to affect the results obtained by heat treatment; the only limitations as to particle size appear to be those imposed by the requirements of the use to which the oxide is put. Satisfactory compressed cores may be made from a finely divided oxide which before incorporation into the cores has particles of such a size that all pass through a No. 40 screen; of course, oxides having all particles of a smaller size may be employed. However, particularly advantageous results are obtained in reducing losses of magnetic bodies by heat treatment of the finely divided oxide when the oxide before heat treatment according to the present invention has particles of such size and shape that the tap density of the oxide is between about .8 to about .85. The tap density is determined by filling a 100 cubic centimeter graduated cylinder with the finely divided oxide, closing the end with a stopper, and tapping the graduate cylinder one hundred times onto a wooden surface from a height of 8 inches; the tap density is the weight of the oxide in grams divided by the volume of the oxide in cubic centimeters.

In accordance with the present invention, such an oxide is advantageously heated in an inert atmosphere to a temperature between about 980° C. and about 1020° C.; preferably it is heated to a temperature of about 1000° C. The time of heating is such as to impart the desired characteristics to the oxide.

The heating may be accomplished in various manners. Advantageously, the finely divided ferroso-ferric oxide is heated while it is in a mass of loose particles before being incorporated into the magnetic body. At the temperatures and under the conditions employed in the process of the invention no, or very little, sintering together of the oxide particles occurs. If desired, a suitable finely divided electrical insulating material capable of resisting the heat treatment temperatures, such as an inorganic ceramic type insulating material comprising talc or kaolin, may be mixed with the mass of loose oxide particles before they are subjected to heat; the presence of the insulating material reduces the possibility of sintering of the oxide particles. The improvement in properties of the ferroso-ferric oxide may also be obtained if the finely divided oxide is heated after it has been formed into a magnetic body either of the compressed type or the loose particle type. However, in general, it is advantageous to heat the particles when they are in loose form with or without an added insulating material since this reduces the possibility of sintering together of the oxide particles, which tends to cause a loss of desirable magnetic characteristics.

The inert atmosphere may be provided in various manners. Thus the ferroso-ferric oxide being treated, either in loose form or otherwise, may be heated in a vacuum or in an inert gas of which nitrogen is an example. However, for maximum ease in carrying out the process and freedom from complexity of the heating apparatus required, it is more desirable to heat the oxide while it is contained in a sealed pot containing substantially no air. The pot should be formed of a material which does not tend to combine with the oxide at the high temperatures employed; for example, a pot formed of a refractory nickel-iron-chro-

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mium alloy metal, with or without an aluminum oxide lining, has been found satisfactory.

The high temperatures employed may be obtained in different ways. For example, the oxide may be heated in an electrical furnace or in a gas-fired furnace, care being taken in each case to insure that the oxide is heated under an inert atmosphere.

The time of heating is not critical. Improvements in the properties of the ferroso-ferric oxide have been obtained when the oxide was heated for as short a time as one-half hour and as long a time as five hours; heating times of longer than five hours may be employed. From the commercial standpoint, of course, shorter heating times are desirable. It has been found that highly advantageous results are obtained when the finely divided oxide is heated from one to one and one-half hours.

The ferroso-ferric oxide may be cooled from the heat treatment temperatures at any suitable rate. In general, it is merely necessary to cool the oxide without any particular precautions to change or control the rate of cooling, provided the oxide is maintained in an inert atmosphere during at least the time it is at a substantially elevated temperature.

The temperature at which best results are obtained when the oxide is heated is quite critical, as is apparent from the curve of Fig. 1. In obtaining the data for this curve, each of several masses of a loose ferroso-ferric oxide powder of the kind described above was respectively heated in a nitrogen atmosphere for one and one-half hours to a different temperature which fell between temperature 700° C. and 1100° C., one of the masses of powder being heated to 1000° C. From each of these masses of powder, without added insulation, a toroidal core of an identical predetermined size was pressed at a pressure of 50 tons per square inch. The quantity ΔQ was then determined for each of said cores.

The quantity Q for an inductance coil is given by the expression

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

where F is the frequency of the current, L is the inductance of the coil and R is its resistance. The quantity ΔQ for a core is equal to the difference between the Q of a coil alone and the Q of the same coil with the magnetic core therein; it thus indicates the magnitude of the losses of the core. A core with a positive ΔQ value or a low negative ΔQ value has low loss characteristics. The quantity ΔQ for each of the cores tested in obtaining the values for the curve of Fig. 1 was determined by means of a Q -meter of the type described on page 15 of the May 1942 issue of the magazine "FM Radio—Electronic Engineering and Design," published by the FM Company, New York city, and by a process generally similar to that described therein. The Q -meter was provided with an inductance coil into which one of the cores could be inserted. The effective Q of the coil without the core was determined as described on said page of said publication at a frequency of 50 megacycles; the effective Q of the coil with the core inserted therein was determined at the same frequency in the same manner; the quantity ΔQ corresponding to the difference in these Q values was an indication of the losses introduced by the core.

As is apparent from Fig. 1, heating of the ferroso-ferric oxide tested to a temperature on

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the order of 1000° C. reduced the core losses substantially and to a much greater extent than did heating of the oxide to temperatures below and above this temperature. The curve of Fig. 1 thus shows that the heating temperature range indicated above, from about 980° C. to about 1020° C. is quite critical in providing maximum improvement in properties.

The above curve is purely illustrative in indicating the effects of temperature of heating of a particular oxide on core losses of a specific type of core; different curves may be obtained when different oxides are employed or when cores of different sizes and shapes are tested. For example, cores having a ΔQ value of over +5 at a frequency of 50 megacycles may be made of oxide treated according to the present invention.

The finely divided ferroso-ferric oxide after treatment according to the present invention possesses permeability and other magnetic characteristics which render the material highly advantageous for use in magnetic bodies, particularly bodies employed in high frequency circuits operating at frequencies between about 25 and about 200 megacycles. The characteristics of the heat treated oxide are such that when formed into magnetic bodies such as cores, the bodies have low loss characteristics, and the loss characteristics and other magnetic properties of the bodies are extremely stable so that they do not change substantially even if the bodies are employed at high temperatures or at fluctuating temperatures.

More specifically, heat treatment, according to the present invention, of finely divided ferroso-ferric oxide of the kind the heat treatment of which is contemplated by the present invention substantially reduces the losses of magnetic bodies formed of the oxide if the oxide originally causes high losses, and does not appreciably harm the stability of the loss characteristics and other magnetic properties of the bodies if the untreated oxide produces bodies possessing such stability; in other cases in which the oxide before heat treatment does not cause high losses, heat treatment according to the present invention does not increase such losses but greatly increases the stability of the loss characteristics and the magnetic properties of the bodies even if they are employed at high or fluctuating temperatures; in still other cases in which the oxide before heat treatment produces magnetic bodies having both high loss characteristics and poor stability of loss characteristics and other magnetic properties, heat treatments of the oxide according to the present invention reduces the losses of the magnetic bodies and greatly improves the stability of the loss characteristics and other magnetic properties even at high or fluctuating temperatures.

The reasons are not definitely known for the improvement of the loss characteristics and other magnetic properties, and the improved stability thereof, of magnetic bodies comprising the finely divided ferroso-ferric oxide heat treated according to the invention. Examination of the oxide before and after heat treatment by means of X-rays has not indicated any substantial recrystallization; the sizes of the particles of the finely divided oxide after heat treatment are substantially the same as the sizes of the particles before treatment.

As has been indicated above, in general, substantially no sintering of the particles of the oxide occurs when the oxide is heated in loose form with or without an added insulating material. Grinding of the heat treated oxide is

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unnecessary; at most a sieving of the heat treated oxide may be desirable.

Ferroso-ferric oxide which has been heat-treated according to the present invention while in a loose mass may be formed into compressed cores with or without separate insulation material. Figure 2 illustrates one form of core embodying oxide heat-treated according to the invention.

In general it is desirable to employ separate insulation material to control or adjust the permeability characteristics of the resulting cores. Various types of organic and inorganic insulating materials may be employed, such as are known to the art. As examples of organic insulating materials may be mentioned thermosetting organic resins such as phenol-formaldehyde condensation products. As examples of suitable inorganic insulating materials may be mentioned the ceramic insulating materials disclosed in Patent 1,943,115, issued January 9, 1934, to W. C. Ellis. The compressed cores may be formed by procedures known to those in the art. For example, the heat-treated finely divided oxide particles may be mixed with and coated with a suitable ceramic insulation in liquid form, dried, and then pressed into cores with or without the addition of another insulating material of the ceramic or organic type. Or the heat-treated oxide particles may be mixed with inorganic or organic insulation and pressed into core shape.

As has been indicated above, ferroso-ferric oxide which has been heat-treated while in a loose mass according to the present invention may be employed in cores of the loose powder type if desired. As was also indicated above, the finely divided ferroso-ferric oxide may be formed into compressed or loose type cores which after formation are heat-treated according to the present invention. If an insulating material is used in such a core, it is necessary to employ one which resists the high temperatures employed, such as a ceramic insulation.

The following is an example of a preferred process of the present invention:

Finely divided ferroso-ferric oxide, sold commercially as a synthetic chemically pure grade, having a molal $\text{Fe}_2\text{O}_3/\text{FeO}$ ratio of 1.4 and containing less than 0.03 per cent metallic iron and only traces of other impurities, all the particles of which oxide passed through a No. 40 screen, and which oxide had a tap density of .81, was placed in loose form in a nickel-iron-chromium alloy metal pot lined with aluminum oxide. The pot was filled with said oxide and was sealed to exclude substantially all air. The sealed pot was then placed in a previously heated furnace and the finely divided oxide was heated at a temperature of about 1000° C., as determined by a thermo-couple, for about one and one-half hours. The pot was removed from the furnace and allowed to cool to room temperature before the seal was broken. Any red oxide found on the surface of the heat treated oxide was removed and discarded. The remaining black oxide was passed through a No. 40 screen. Cores of the form shown in Fig. 2 and pressed from the heat-treated powder at a pressure of 50 tons per square inch at frequencies on the order of 50 megacycles had loss characteristics only about one-fifth as great as those of identical cores pressed from the identical powder which was not heat treated, the loss characteristics being determined as described above in connection with Fig. 1.

While the invention has been primarily dis-

cussed above in connection with the manufacture of magnetic cores for inductance coils, it is apparent that the invention may be employed in the treatment of ferroso-ferric oxide powder for other magnetic purposes. Various modifications may be made in the methods of heating, and in other features of the process described above. It is intended that the patent shall cover by suitable expression of the appended claims whatever features of patentable novelty reside in the invention.

What is claimed is:

1. A method of improving the properties of a ferroso-ferric oxide rendering it suitable for use in a magnetic body comprising heating a finely divided ferroso-ferric oxide comprising a substantial molecular excess of Fe_2O_3 with respect to FeO and being substantially free of impurities, to a temperature in excess of 700°C . in an inert atmosphere for a period of at least about one-half hour.

2. A method of improving the properties of a ferroso-ferric oxide rendering it suitable for use in a magnetic body comprising heating a finely divided ferroso-ferric oxide comprising a substantial molecular excess of Fe_2O_3 with respect to FeO and being substantially free of impurities, to a temperature between about 980°C . and about 1020°C . in an inert atmosphere for at least about one-half hour.

3. A method of improving the properties of a ferroso-ferric oxide rendering it suitable for use in a magnetic body to be employed in a circuit operating at a frequency of at least about 25 megacycles comprising heating a finely divided ferroso-ferric oxide comprising a molecular proportion of Fe_2O_3 between about 1.25 and 1.75 times as great as the molecular proportion of FeO and containing less than about 0.03 per cent by weight of metallic iron and substantially no other impurities, to a temperature between about 980°C . and about 1020°C . in an inert atmosphere for at least about one-half hour.

4. A method of improving the properties of a ferroso-ferric oxide rendering it suitable for use in a magnetic body comprising heating a loose mass of a finely divided ferroso-ferric oxide comprising a substantial molecular excess of Fe_2O_3 with respect to FeO and being substantially free of impurities, to a temperature between about 980°C . and about 1020°C . in an inert atmosphere for a period of at least about one-half hour.

5. A method of improving the properties of a ferroso-ferric oxide rendering it suitable for use in a magnetic body comprising heating a loose mass of a finely divided ferroso-ferric oxide comprising a substantial molecular excess of Fe_2O_3 with respect to FeO and being substantially free of impurities, to a temperature between about 980°C . and about 1020°C . in an atmosphere of an inert gas for a period of at least about one-half hour.

6. A method of improving the properties of a ferroso-ferric oxide rendering it suitable for use in a magnetic body comprising heating a loose mass of a finely divided ferroso-ferric oxide comprising a substantial molecular excess of Fe_2O_3 with respect to FeO and being substantially free of impurities, to a temperature between about 980°C . and about 1020°C . in a substantially evacuated container for a period of at least about one-half hour.

7. A method of improving the properties of a

ferroso-ferric oxide rendering it suitable for use in a magnetic body comprising heating a loose mass of a finely divided ferroso-ferric oxide comprising a substantial molecular excess of Fe_2O_3 with respect to FeO and being substantially free of impurities, to a temperature between about 980°C . and about 1020°C . in an atmosphere of an inert gas for from about one-half hour to about five hours.

8. A method of improving the properties of a ferroso-ferric oxide rendering it suitable for use in a magnetic body comprising heating a loose mass of a finely divided ferroso-ferric oxide comprising a substantial molecular excess of Fe_2O_3 with respect to FeO and being substantially free of impurities, to a temperature between about 980°C . and about 1020°C . in a sealed pot containing a substantially inert atmosphere for from about one-half hour to about five hours.

9. A method of improving the properties of a ferroso-ferric oxide rendering it suitable for use in magnetic bodies comprising heating a loose mass of a finely divided ferroso-ferric oxide comprising a molecular proportion of Fe_2O_3 between about 1.40 to 1.55 times as great as the molecular proportion of FeO and containing less than about 0.03 per cent by weight of metallic iron and substantially no other impurities, and having a tap density between about .8 and .85 to a temperature between about 980°C . and about 1020°C . in an inert atmosphere for at least about one-half hour.

10. The method of improving the properties of magnetic bodies comprising heating a pressed magnetic body comprising a finely divided ferroso-ferric oxide comprising a substantial molecular excess of Fe_2O_3 with respect to FeO and being substantially free of impurities, to a temperature between about 980°C . and about 1020°C . in an inert atmosphere for a period of at least about one-half hour.

11. The method of improving the properties of magnetic cores comprising heating a pressed magnetic core comprising a finely divided ferroso-ferric oxide comprising a molecular proportion of Fe_2O_3 between about 1.40 to 1.55 times as great as the molecular proportion of FeO and containing less than about 0.03 per cent by weight of metallic iron and substantially no other impurities, and having a tap density between about .8 and about .85 to a temperature between about 980°C . and about 1020°C . in an inert atmosphere for at least about five hours.

12. Process of claim 11 in which said inert atmosphere is an atmosphere of an inert gas surrounding said core.

13. Process of claim 11 in which said core is heated in a substantially evacuated container.

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