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PROCESS OF MANUFACTURING A MAGNETIC
MATERIAL AND MAGNETIC CORE
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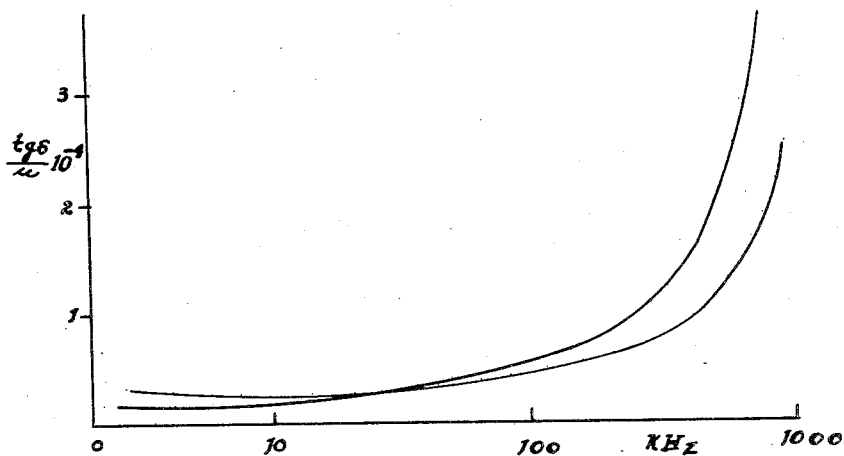


FIG. 1

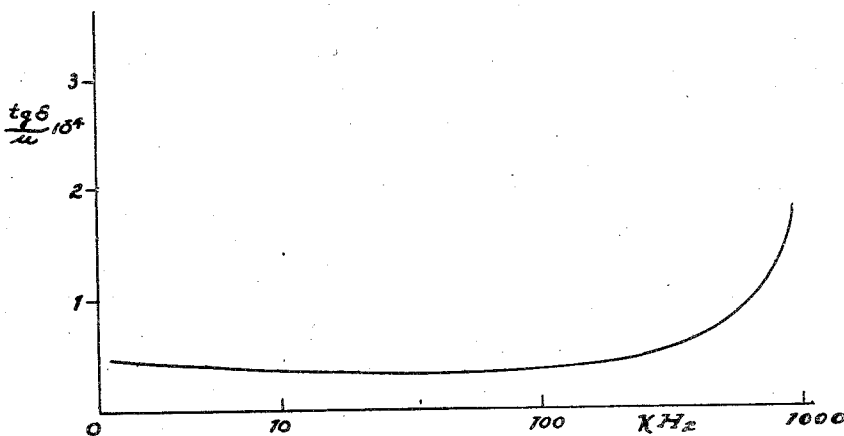


FIG. 2

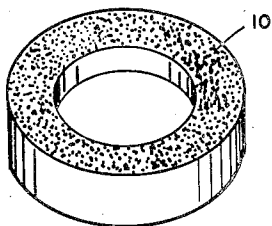


FIG. 3

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UNITED STATES PATENT OFFICE

2,452,531

PROCESS OF MANUFACTURING A MAGNETIC MATERIAL AND MAGNETIC CORE

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Section 1, Public Law 690, August 8, 1946
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19 Claims. (Cl. 252—62.5)

1

This invention relates to the manufacture of magnetic cores having a cubic ferrite as a magnetic material, which give rise to small losses at low inductance, even with high frequencies, more particularly cores for telegraphy and telephony purposes, for example for filter coils, pupin coils, etc.

It is known that magnetic cubic ferrites ordinarily have a high specific electrical resistance, for example 1000 ohms cm. and higher, and since with a resistance of 10 to 100 ohms cm. the eddy-currents are already extremely feeble, such ferrites practically do not exhibit eddy-current losses. It appears, however, that in spite of the eddy-current losses being substantially nil, there may nevertheless occur considerable losses.

Reference is made to my copending U. S. applications Serial Nos. 616,928 filed September 17, 1945, and 617,392 filed September 19, 1945, of which the present application is a continuation-in-part.

The present invention is based on the recognition of the fact that these losses are connected with the oxygen contents of ferrite.

With regard to the percentage of oxygen it may be mentioned that it is known that a ferrite when heated to high temperatures such, for example, as are utilized in its preparation, may split off oxygen. In order to avoid such a deficiency of oxygen, the heating in question was carried out in pure oxygen.

Now, it has been found that, even if a heating required for the preparation or any other purpose is carried out in pure oxygen, a small deficiency of oxygen frequently occurs and that to our surprise such a small deficiency of oxygen, which may be only a few hundredths of percent by weight of the total weight of the ferrite, is highly disadvantageous for the losses.

Now, the invention consists in that in the manufacture of a magnetic material constituted by a magnetic cubic ferrite having a high specific resistance, higher than 1000 ohms cm., so high a percentage of oxygen of the ferrite is provided for that the loss factor $tg\delta$ in the frequency range of from 10 to 100 kilocycles/sec. is less than 0.06. The value $tg\delta$ is equal to

$$\frac{R}{\omega L}$$

R being the loss resistance measured whilst avoiding the occurrence of dielectric losses and deduction being made of the direct-current resistance, L representing the inductance of a coil

2

wound on an annular core of the ferrite, and ω being the annular frequency.

A percentage of oxygen according to the invention may be obtained in different manners, according to the kind and the composition of the ferrite.

The process, fundamentally consists simply that by a suitable choice of the conditions care is taken that the heating temperature required for the preparation of the ferrite is maintained sufficiently low.

With regard to the heating temperature it is observed that, for example, with a ferrite which is prepared by heating an intimate mixture of the solid oxides constituting the ferrite the temperature will depend on the intensity with which, and the fineness to which the mixture is ground. A very fine mixture ground for a long time will within a reasonable period be capable of yielding a homogeneous product thoroughly reacted at lower temperatures than a coarser mixture ground for a shorter time; as will be explained more fully hereinafter, such a complete reaction is of great importance in view of the initial permeability of the final product.

Further, it has been found that in definite cases, the sintering temperature is somewhat decreased by decreasing the percentage of iron oxide of the ferrite.

Although the variation of the conditions under which a ferrite is prepared permits a certain amount of latitude in the heating temperature required, it is frequently not possible in practice to provide for a sufficiently high percentage of oxygen already during the preparation of the ferrite. According to the invention, such a ferrite having a too low percentage of oxygen, which consequently is unsaturated with respect to oxygen, may be caused to absorb oxygen, for example at lower temperature.

The conditions, more particularly the temperature at which a ferrite is caused to absorb oxygen, depend on several factors, such as on the amount of oxygen which must be absorbed to obtain a sufficiently low value for $tg\delta$, on the degree to which all parts of the ferrite are accessible to oxygen, on its fineness of grain and hence on its porosity, and further on the kind and the composition of the ferrite used.

It has been found that with unvaried oxygen pressure the amount of oxygen absorbed increases with a decrease of temperature. On the other hand it is necessary to consider the circumstance that the speed of absorption of oxygen decreases with a decrease in temperature. This

speed further strongly depends on the fineness of grain and the porosity of the ferrite so that in view of the time which would otherwise be taken up by the absorption of oxygen, it is desirable that the ferrite should be utilized in the fine-grained porous state.

It is observed that to our surprise it was found possible to manufacture a ferrite in a sintered state which is sufficiently compact to serve as a magnetic core whilst maintaining at the same time sufficient porosity.

An important factor which must be considered in utilizing the invention is the initial permeability of the final product since the serviceability of a magnetic core is substantially determined by the value of the quotient

$$\frac{tg\delta}{\mu}$$

of the above-defined loss factor $tg\delta$ and the initial permeability μ measured on an annular core. Cores having a high initial permeability and low losses are of great importance. Consequently, in the manufacture of such cores one will desire to combine the steps for obtaining a sufficiently high percentage of oxygen with steps which are required to obtain a high initial permeability, care having to be taken to see to it that the different steps do not come into conflict with one another.

It has now been found that for obtaining a high value for the initial permeability it is desirable to ensure that the ultimate ferrite approaches as far as possible the state of a single homogeneous phase, i. e. it must be ensured that the ferrite forming initial mixture reacts thoroughly while during cooling the ferrite once produced must be prevented as far as possible from splitting off a second phase. The latter case may occur when during cooling the ferrite splits off one of its constitutive oxides which at high temperature is so-to-say maintained in an oversaturated solid solution, or when during cooling the ferrite will fall into its constitutive oxides.

If there is a danger of a second phase being separated, this may be avoided by rapid cooling, although a rapid cooling, more rapid than, for example, at the rate of 10° C. per minute is as a rule to be avoided, since thus chilling tensions may result which are detrimental to the permeability. The most advantageous speed of cooling may be easily ascertained experimentally in each individual case. From the foregoing it further follows that during the absorption of oxygen the temperature must preferably remain above the temperature at which a second phase may be produced.

Further it has been found that, like with other magnetic materials, the initial permeability of the ferrite is at a maximum at a temperature in the neighborhood of, but less than the Curie point, the Curie point being the temperature at which the magnetic material for all practical purposes may be considered non-magnetic; in view thereof a ferrite having a Curie-point located between 40° C. and 250° C. is highly advantageous. A ferrite of this kind is obtained, for example, by combining zinc ferrite, which has a low Curie-point, with one or more ferrites having a higher Curie-point, such as nickel ferrite, so that a mixed crystal is obtained.

It is also possible to influence the Curie-point of the ferrite by regulating its percentage of iron oxide while, in addition, the percentage of oxygen may be of influence. The term Curie-point

has to be understood in this case to mean the temperature at which the ferrite, as regards its initial permeability, changes into a state to be considered as unmagnetic for practical purposes.

For obtaining a high initial permeability the use of pure raw materials also is of much importance.

As previously mentioned, there is a danger of the steps required for the obtainment of a high initial permeability coming into conflict with steps required for a sufficiently high percentage of oxygen which is desirable in view of the losses. Thus, for obtaining a high initial permeability, heating to a high temperature is generally desirable in order to facilitate the thorough reaction and the formation of a homogeneous phase. The heating temperature necessary to obtain the highest possible initial permeability is in most cases, however, such that the percentage of oxygen and the possibility of absorption of oxygen are detrimentally affected by such a heating, the obtainment of low losses thus being rendered difficult. Consequently, in this case one has to arrive at a compromise solution.

In my copending U. S. application Serial No. 616,928, filed September 17, 1945, I have described a method of manufacturing a homogeneous mixed crystal ferrite having an oxygen content at which the coefficient of losses $tg\delta$ in a frequency range between 10 kc./sec. and 1000 kc./sec. is less than 0.06 which is particularly suited for radio purposes. In order to provide for such a percentage of oxygen, it is necessary in the preparation of ferrite, or with a heating for other purposes not to utilize an excessive temperature; due to the avoidance of a high temperature an optimum value for the permeability is frequently not obtained. In this case one has to do with a compromise, as above referred to.

Now, it has been found that, when using a higher sintering temperature, it is still possible to obtain such a percentage of oxygen that the loss factor $tg\delta$ through a region extending to less high frequencies is lower than 0.06. With higher frequencies the loss factor exceeds this limit.

This material sintered at a higher temperature differentiates from the material sintered at a lower temperature amongst others by a coarser granular structure.

Such a magnetic material which has a loss factor higher than 0.06 with frequencies lower than 1000 kilocycles/sec. is not suited to radio purposes, it is true, but it is still well serviceable for telegraphy or telephony. Besides, it offers the advantage that in many cases, due to the higher permissible sintering temperature, a higher initial permeability is obtained.

Although, as a matter of fact, the initial permeabilities that can be obtained are different, amongst others because one ferrite during cooling separates a second phase less easily than another and may thus be brought more easily into the state which approaches that of a single homogeneous phase, with the use of a ferrite of suitable composition it has in many cases been found possible to make a magnetic material of which the value of

$$\frac{tg\delta}{\mu}$$

is smaller than 0.0001 from 10 to 100 kilocycles/sec. Such a material is excellently suited to telegraphy and telephony purposes, for example to the manufacture of filter coils, with which it is desired to utilize frequencies of about 10 to

5

100 kilocycles/sec. Also for pupin coils which as a rule are used with frequencies of from 300 to 2000 cycles/sec., such a magnetic material is excellently suited. According to the invention, very good results may be obtained with ferrites of which the percentage of iron oxide is less than 50 mol. per cent.

As previously mentioned, the serviceability of a magnetic material is substantially determined by the value of the quotient

$$\frac{tg\delta}{\mu}$$

To explain this, it is observed that the reason why this material constant has such a suitable value for the judgment of a magnetic material is that in a magnetic circuit having one or more so-called air-gaps the quotient

$$\frac{tg\delta_{eff}}{\mu_{eff}}$$

of the effective loss factor $tg\delta_{eff}$ and of the effective initial permeability μ_{eff} with constant magnetic load of the material is independent of the number and the size of the air-gaps and hence equal to

$$\frac{tg\delta}{\mu}$$

in which $tg\delta$ and μ are measured on an annular core.

When the constant value of the quotient for a ferrite has once been fixed, for example by measurements on an annular core of the ferrite, the loss factor of another core built up from this material may consequently be found by multiplication of the constant with the effective permeability of this core.

A ferrite according to the invention is preferably prepared by compressing and subsequently sintering a mixture of the oxides constituting the ferrite, or a corresponding mixture of compounds which upon heating change into oxide.

To obtain a product reacted as thoroughly as possible, it is desirable, as previously mentioned, to start from a ferrite-forming initial mixture of great fineness and reactivity. In order to obtain a great reactivity, the initial mixture may be ground for a long time and with great intensity, this grinding being preferably effected to such extent that an average size of particles smaller than 1μ is obtained.

It is alternatively possible to precipitate a solution containing all the metals constituting the ferrite with the aid of a base and to dry the deposit obtained, which may in part already exhibit ferrite structure. Subsequently, in order to improve the mouldability, the deposit is preferably heated to 500 to 700° C.

Further, in the preparation of a ferrite it is possible to utilize sintering several times, that is to say that the mixture at first sintered is pulverized and sintered again. In this case the first sinterings are effected at lower temperature, during which the mixture does not yet show a complete reaction. The product obtained can then be easily reground to great fineness. A compression of the mixture to be preliminarily sintered is preferably omitted, also to facilitate grinding. This method of preparation offers the advantage that at least a well reacted product is obtained, which increases the value of the initial permeability.

In my copending U. S. application Serial No. 616,928, filed September 17, 1945, of which the

6

present application is a continuation-in-part, I have described and claimed homogeneous mixed crystal ferrites having an oxygen content at which the coefficient of losses $tg\delta$ in the frequency range between 10 kc./sec. and 1000 kc./sec. is less than 0.06. Cores having such magnetic materials are excluded from the exclusive rights now claimed. It is still mentioned that the expression magnetic core in the present invention covers not only a core arranged inside a coil but as a rule parts of electro-magnetic constructions which are utilized in view of their magnetic properties, for example also parts for magnetic screening.

In order that the invention may be more readily understood it will now be described with reference to the following examples and the accompanying drawing in which:

Fig. 1 shows the relationship between the quotient of the coefficient of losses and permeability

$$\frac{tg\delta}{\mu}$$

with frequency for Example I.

Fig. 2 shows the relationship between the quotient of the coefficient of losses and permeability

$$\frac{tg\delta}{\mu}$$

with frequency for Example II; and

Fig. 3 shows a core composed essentially of magnetic ferrite material according to the invention.

Referring to the drawing, I have shown a sample ring core 10 of a homogeneous mixed crystal ferrite which is provided with an oxygen content at which the coefficient of losses $tg\delta$ is less than 0.06 in a frequency range between 10 and 100 kc./sec. and greater than 0.06 at a frequency between 100 and 1000 kc./sec.

Example I

An intimate mixture of pure magnesium oxide, zinc oxide and iron oxide in a molecular ratio of 26.5:26.5:47 is ground during 3 hours in an iron hurling mill. A ring of 3 cm. in diameter and 4 x 4 mm. in section is moulded from the mixture under a pressure of 4 tons/cm.² with the use of water as a plastifying and binding agent. This ring is heated in oxygen to 1300° C. during 1 hour, followed by cooling in oxygen at a rate of about 3° C. per minute. The initial permeability amounts to 350. The values of

$$\frac{tg\delta}{\mu}$$

are plotted in Figure 1 in dependence on frequency, curve *a*. From this curve it follows that with frequencies higher than 800 kilocycles/sec. the value for $tg\delta$ becomes higher than 0.06.

If the heating is effected at 1400° C. instead of 1300° C., one obtains the values of curve *b* shown in Figure 1. In this case the initial permeability amounts to 525. With frequencies higher than 350 kilocycles/sec. $tg\delta$ becomes higher than 0.06.

Example II

A mixture of technical copper oxide, zinc oxide and iron oxide, in a molecular ratio of

20.7:31.6:47.7

calculated on the pure oxides, which is added by 1% by weight of brownstone, is ground during 3 hours and subsequently moulded to form a ring in the manner described in Example I.

This ring is sintered in oxygen at 1050° C. during 1 hour and subsequently slowly cooled down

to 600° C., which temperature is maintained during 14 hours, all this in oxygen, whereafter further cooling takes place. The copper-zinc-ferrite core has an initial permeability of 385. The values of

$$\frac{tg\delta}{\mu}$$

are plotted in Figure 2 in dependence of frequency. From this figure it follows that with frequencies higher than 900 kilocycles/sec. the value for $tg\delta$ becomes higher than 0.06.

For the purpose of defining the terms "ferrite," "mixed crystal," and "soft" magnetic materials, the following definitions will be employed in connection with the above-noted terms as used throughout the specification and in the appended claims.

A "ferrite" is a crystalline material which is a compound of the reaction product of a metal oxide and iron oxide having the empirical formula MFe_2O_4 wherein M represents a bivalent metal. This material may also be defined as a metallic salt of the hypothetical acid $H_2Fe_2O_4$.

A "mixed crystal" ferrite is a ferrite material comprising two or more ferrites as hereinbefore defined which are chemically combined together to form a single homogeneous crystalline compound.

The term "soft" magnetic material means magnetic material having a low remanence and a low coercivity when the applied inductive field is removed from the material.

Within the scope of the definitions noted above, I have described my invention with specific examples and methods of execution, which, however, will suggest other obvious modifications to those skilled in the art without departing from the spirit and scope of my invention.

What I claim is:

1. A soft ferromagnetic core material having low magnetic losses in a frequency range between 10 kc./sec. and 100 kc./sec. and increasing magnetic losses above 100 kc./sec. and which is particularly suited for cores, inductance coils and the like employed in telephony and telegraphy circuits operating within said range of frequencies, consisting essentially of a cubic homogeneous mixed crystal ferrite constituted by a plurality of ferrites, said mixed crystal ferrite having a specific resistivity greater than about 1000 ohm-cm. and having an oxygen content at which the coefficient of losses $tg\delta$ thereof is less than about 0.06 in the range of frequencies between about 10 kc./sec. and 100 kc./sec. and greater than 0.06 at a frequency between about 100 kc./sec. and about 1000 kc./sec.

2. A soft ferromagnetic core material having low magnetic losses in a frequency range between 10 kc./sec. and 100 kc./sec. and increasing magnetic losses above 100 kc./sec. and which is particularly suited for cores, inductance coils and the like employed in telephony and telegraphy circuits operating within said range of frequencies, consisting essentially of a cubic homogeneous mixed crystal ferrite constituted by a plurality of ferrites, said mixed crystal ferrite having a specific resistivity greater than about 1000 ohm-cm. and having an oxygen content at which the coefficient of losses $tg\delta$ of said mixed crystal ferrite is less than about 0.06 in the range of frequencies between about 10 kc./sec. and 100 kc./sec. and greater than 0.06 at a frequency between about 100 kc./sec. and about 1000 kc./sec., said mixed crystal ferrite having a Curie point between 40° C. and 250° C.

3. A soft ferromagnetic core material having low magnetic losses in a frequency range between 10 kc./sec. and 100 kc./sec. and increasing magnetic losses above 100 kc./sec. and which is particularly suited for cores, inductance coils and the like employed in telephony and telegraphy circuits operating within said range of frequencies, consisting essentially of a cubic homogeneous mixed crystal ferrite constituted by zinc ferrite and a second ferrite having a Curie point greater than that of said zinc ferrite, said mixed crystal ferrite having a specific resistivity greater than about 1000 ohm-cm. and an oxygen content at which the coefficient of losses $tg\delta$ of said mixed crystal ferrite is less than about 0.06 in the range of frequencies between about 10 kc./sec. and 100 kc./sec. and greater than 0.06 at a frequency between about 100 kc./sec. and about 1000 kc./sec., said mixed crystal ferrite having a Curie point between about 40° C. and 250° C.

4. A soft ferromagnetic core material having low magnetic losses in a frequency range between 10 kc./sec. and 100 kc./sec. and increasing magnetic losses above 100 kc./sec. and which is particularly suited for cores, inductance coils and the like employed in telephony and telegraphy circuits operating within said range of frequencies, consisting essentially of a cubic homogeneous mixed crystal ferrite constituted by zinc ferrite and a second ferrite having a Curie point greater than that of said zinc ferrite, said mixed crystal ferrite having a specific resistivity greater than about 1000 ohm-cm. and an oxygen content at which the quotient of the coefficient of losses $tg\delta$ divided by the initial permeability μ of said mixed crystal ferrite in the frequency range between about 10 kc./sec. and 100 kc./sec. is less than about 0.0001 and having a loss factor $tg\delta$ which is greater than 0.06 at a frequency between about 100 kc./sec. and about 1000 kc./sec., said mixed crystal ferrite having a Curie point between about 40° C. and 250° C.

5. A soft ferromagnetic core material having low magnetic losses in a frequency range between 10 kc./sec. and 100 kc./sec. and increasing magnetic losses above 100 kc./sec. and which is particularly suited for cores, inductance coils and the like employed in telephony and telegraphy circuits operating within said range of frequencies, consisting essentially of a cubic homogeneous mixed crystal ferrite constituted by zinc ferrite and magnesium ferrite, said mixed crystal ferrite having a specific resistivity greater than about 1000 ohm-cm. and an oxygen content at which the quotient of the coefficient of losses $tg\delta$ divided by the initial permeability μ of said mixed crystal ferrite in the frequency range between about 10 kc./sec. and 100 kc./sec. is less than about 0.0001 and having a loss factor $tg\delta$ which is greater than 0.06 at a frequency between about 100 kc./sec. and about 1000 kc./sec., said mixed crystal ferrite having a Curie point between about 40° C. and 250° C.

6. A soft ferromagnetic core material having low magnetic losses in a frequency range between 10 kc./sec. and 100 kc./sec. and increasing magnetic losses above 100 kc./sec. and which is particularly suited for cores, inductance coils and the like employed in telephony and telegraphy circuits operating within said range of frequencies, consisting essentially of a cubic homogeneous mixed crystal ferrite constituted by zinc ferrite and copper ferrite, said mixed crystal ferrite having a specific resistivity greater than about 1000 ohm-cm. and an oxygen content at

which the quotient of the coefficient of losses $tg\delta$ divided by the initial permeability μ of said mixed crystal ferrite in the frequency range between about 10 kc./sec. and 100 kc./sec. is less than about 0.0001 and having a loss factor $tg\delta$ which is greater than 0.06 at a frequency between about 100 kc./sec. and about 1000 kc./sec., said mixed crystal ferrite having a Curie point between about 40° C. and 250° C.

7. A soft ferromagnetic core material having low magnetic losses in a frequency range between 10 kc./sec. and 100 kc./sec. and increasing magnetic losses above 100 kc./sec. and which is particularly suited for cores, inductance coils and the like employed in telephony and telegraphy circuits operating within said range of frequencies, consisting essentially of a cubic homogeneous mixed crystal ferrite constituted by magnesium ferrite and zinc ferrite having oxide components in an amount equivalent to approximately 26.5 mol. per cent of magnesium oxide, 26.5 mol. per cent of zinc oxide, and 47 mol. per cent of iron oxide, said mixed crystal ferrite having a specific resistivity greater than about 1000 ohm-cm. and an oxygen content at which the quotient of the coefficient of losses $tg\delta$ divided by the initial permeability μ of said material in the frequency range between about 10 kc./sec. and 100 kc./sec. is less than about 0.0001 and having a loss factor $tg\delta$ which is greater than 0.06 at a frequency between about 100 kc./sec. and about 1000 kc./sec., said mixed crystal ferrite having a Curie point between about 40° and 250° C.

8. A soft ferromagnetic core material having low magnetic losses in a frequency range between 10 kc./sec. and 100 kc./sec. and increasing magnetic losses above 100 kc./sec. and which is particularly suited for cores, inductance coils and the like employed in telephony and telegraphy circuits operating within said range of frequencies, consisting essentially of a cubic homogeneous mixed crystal ferrite constituted by copper ferrite and zinc ferrite having oxide components in an amount equivalent to approximately 20.7 mol. per cent of copper oxide, 31.6 mol. per cent of zinc oxide, and 47.7 mol. per cent of iron oxide, said mixed crystal ferrite having a specific resistivity greater than about 1000 ohm-cm. and an oxygen content at which the quotient of the coefficient of losses $tg\delta$ divided by the initial permeability μ of said material in the frequency range between about 10 kc./sec. and 100 kc./sec. is less than about 0.0001 and having a loss factor $tg\delta$ which is greater than 0.06 at a frequency between about 100 kc./sec. and about 1000 kc./sec., said mixed crystal ferrite having a Curie point between about 40° C. and 250° C.

9. The method of manufacturing a soft ferromagnetic core material having low magnetic losses in a frequency range between 10 kc./sec. and 100 kc./sec. and increasing magnetic losses above 100 kc./sec. which is particularly suited for cores, inductance coils and the like employed in telephony and telegraphy circuits operating within said range of frequencies, comprising the steps of heating a mixture of a first cubic ferrite and a second cubic ferrite to a temperature between about 1000° C. and about 1400° C. in an oxygen-controlling atmosphere to produce a homogeneous mixed crystal ferrite, and regulating the oxygen content in said mixed crystal ferrite to produce a mixed crystal ferrite material having a coefficient of losses $tg\delta$ which is less than about 0.06 in the range of frequencies between

about 10 kc./sec. and 100 kc./sec. and greater than 0.06 at a frequency between about 100 kc./sec. and about 1000 kc./sec.

10. The method of manufacturing a soft ferromagnetic core material having low magnetic losses in a frequency range between 10 kc./sec. and 100 kc./sec. and increasing magnetic losses above 100 kc./sec. which is particularly suited for cores, inductance coils and the like employed in telephony and telegraphy circuits operating within said range of frequencies, comprising the steps of heating a mixture of a first cubic ferrite with a second cubic ferrite in an oxygen atmosphere to a temperature between about 1000° C. and about 1400° C. to produce a homogeneous mixed crystal ferrite, and regulating the oxygen content in said mixed crystal ferrite to produce a mixed crystal ferrite material having a coefficient of losses $tg\delta$ which is less than about 0.06 in a range of frequencies between about 10 kc./sec. and 1000 kc./sec. and greater than 0.06 at a frequency between about 100 kc./sec. and about 1000 kc./sec.

11. The method of manufacturing a soft ferromagnetic core material having low magnetic losses in a frequency range between 10 kc./sec. and 100 kc./sec. and increasing magnetic losses above 100 kc./sec. which is particularly suited for cores, inductance coils and the like employed in telephony and telegraphy circuits operating within said range of frequencies, comprising the steps of pulverizing a mixture of a first cubic ferrite and a second cubic ferrite to an average grain size of less than about 1 micron, heating the said mixture to a temperature between about 1000° C. and about 1400° C. in an oxygen atmosphere to produce a homogeneous mixed crystal ferrite, regulating the oxygen content in said mixed crystal ferrite to produce a mixed crystal ferrite having a coefficient of losses $tg\delta$ which is less than about 0.06 in a frequency range between about 10 kc./sec. and 100 kc./sec. and greater than 0.06 at a frequency between about 100 kc./sec. and about 1000 kc./sec., and cooling the mixed crystal ferrite material at a temperature rate less than about 10° C. per minute in an oxygen atmosphere to maintain the said oxygen content in said material.

12. The method of manufacturing a soft ferromagnetic material having low magnetic losses in a frequency range between 10 kc./sec. and 100 kc./sec. and increasing magnetic losses above 100 kc./sec. which is particularly suited for cores, inductance coils and the like employed in telephony and telegraphy circuits operating within said range of frequencies, comprising the steps of heating a mixture of a first cubic ferrite and a second cubic ferrite to a temperature less than 1000° C. in an oxygen atmosphere, pulverizing the so heated mixture to a fine grain size, heating the pulverized mixture to a temperature between about 1000° C. and about 1400° C. in an oxygen atmosphere to produce a homogeneous mixed crystal ferrite, regulating the oxygen content of said mixed crystal ferrite to produce a mixed crystal ferrite material having a coefficient of losses $tg\delta$ which is less than about 0.06 in a frequency range between about 10 kc./sec. and 100 kc./sec. and greater than 0.06 at a frequency between about 100 kc./sec. and about 1000 kc./sec., and cooling said mixed crystal ferrite material at a temperature rate less than 10° C. per minute in an oxygen atmosphere to maintain the said oxygen content in said material.

13. The method of manufacturing a soft ferro-

magnetic core material having low magnetic losses in a frequency range between 10 kc./sec. and 100 kc./sec. and increasing magnetic losses above 100 kc./sec. which is particularly suited for cores, inductance coils and the like employed in telephony and telegraphy circuits operating within said range of frequencies, comprising the steps of heating a mixture of zinc ferrite and a second ferrite having a Curie point greater than that of said zinc ferrite in a proportion to produce a homogeneous mixed crystal ferrite having a Curie point between 40° C. and 250° C. to a temperature less than 1000° C. in an oxygen atmosphere, pulverizing the so heated mixture to a fine grain size, heating the pulverized mixture to a temperature between about 1000° C. and 1400° C. in an oxygen atmosphere to produce a homogeneous mixed crystal ferrite, regulating the oxygen content of said mixed crystal ferrite to produce a mixed crystal ferrite material having a coefficient of losses $tg \delta$ which is less than about 0.06 in a frequency range between about 10 kc./sec. and 100 kc./sec. and greater than 0.06 at a frequency between about 100 kc./sec. and about 1000 kc./sec., and cooling said mixed crystal ferrite material in an oxygen atmosphere at a temperature rate less than about 10° C. per minute to maintain the said oxygen content and to obtain a homogeneous mixed crystal ferrite having a Curie point between 40° C. and 250° C.

14. The method of manufacturing a soft ferromagnetic core material having low magnetic losses in a frequency range between 10 kc./sec. and 100 kc./sec. and increasing magnetic losses above 100 kc./sec. which is particularly suited for cores, inductance coils and the like employed in telephony and telegraphy circuits operating within said range of frequencies, comprising the steps of heating a mixture of zinc ferrite and magnesium ferrite in an oxygen atmosphere to a temperature less than 1000° C., pulverizing the so heated mixture to a fine grain size, heating the pulverized mixture to a temperature of about 1300° C. in an oxygen atmosphere to produce a homogeneous mixed crystal ferrite, regulating the oxygen content of said mixed crystal ferrite to produce a mixed crystal ferrite material having a coefficient of losses $tg \delta$ which is less than about 0.06 in a frequency range between about 10 kc./sec. and 100 kc./sec. and greater than 0.06 at a frequency between about 100 kc./sec. and about 1000 kc./sec., and cooling the mixed crystal ferrite material at a temperature rate less than about 10° C. per minute to maintain the said oxygen content in said material.

15. The method of manufacturing a soft ferromagnetic core material having low magnetic losses in a frequency range between 10 kc./sec. and 100 kc./sec. and increasing magnetic losses above 100 kc./sec. which is particularly suited for cores, inductance coils and the like employed in telephony and telegraphy circuits operating within said range of frequencies, comprising the steps of heating a mixture of zinc ferrite and copper ferrite in an oxygen atmosphere to a temperature less than 1000° C., pulverizing the so heated mixture to a fine grain size, heating the pulverized mixture to a temperature of about 1050° C. in an oxygen atmosphere to produce a homogeneous mixed crystal ferrite, regulating the oxygen content of said mixed crystal ferrite to produce a mixed crystal ferrite having a coefficient of losses $tg \delta$ of the mixed crystal ferrite which is less than about 0.06 in a frequency range between about 10 kc./sec. and 100 kc./sec.

and greater than 0.06 at a frequency between about 100 kc./sec. and about 1000 kc./sec. and cooling the mixed crystal ferrite material at a temperature rate less than about 10° C. per minute to maintain the said oxygen content in said material.

16. The method of manufacturing magnetic core material, comprising the steps of intimately mixing pure powdered magnesium oxide, pure powdered zinc oxide, and pure powdered iron oxide in the molecular ratio of 26.5:26.5:5:47, pulverizing said intimate mixture to a grain size smaller than about 1 micron, compressing said mixture into a core under a pressure of approximately 4 tons/cm.², heating said core to a temperature of about 1300° C. for about one hour in an oxygen atmosphere, and cooling said core at a temperature rate of about 3° C. per min. in an atmosphere of pure oxygen to allow said heated core to absorb oxygen so that the coefficient of losses $tg \delta$ is less than about 0.06 at frequencies greater than about 10 kc./sec. and less than about 100 kc./sec. and the initial permeability is about 350.

17. The method of manufacturing magnetic core material, comprising the steps of intimately mixing pure powdered magnesium oxide, pure powdered zinc oxide, and pure powdered iron oxide in the molecular ratio of 26.5:26.5:47, pulverizing said intimate mixture to a grain size smaller than about 1 micron, compressing said mixture into a core under a pressure of approximately 4 tons/cm.², heating said core to a temperature of about 1400° C. for about one hour in an oxygen atmosphere, and cooling said core at a temperature rate of about 3° C. per min. in an atmosphere of pure oxygen to allow said heated core to absorb oxygen so that the coefficient of losses $tg \delta$ is less than about 0.06 at frequencies greater than about 10 kc./sec. and less than about 100 kc./sec. and the initial permeability is about 525.

18. The method of manufacturing magnetic core material, comprising the steps of intimately mixing pure powdered copper oxide, pure powdered zinc oxide, and pure powdered iron oxide in the molecular ratio of 20.7:31.6:47.7, pulverizing said mixture to a grain size smaller than about 1 micron, compressing said mixture into a core under a pressure of about 4 tons/cm.², sintering said core in an oxygen atmosphere at a temperature of about 1050° for approximately one hour, slowly cooling said core in an oxygen atmosphere to a temperature of about 600° C., maintaining the core at about 600° C. for approximately 14 hours in an oxygen atmosphere, and slowly cooling said core in an oxygen atmosphere so that the quotient of the coefficient of losses

$$\frac{tg \delta}{\mu}$$

is less than about 0.0001 at frequencies between about 10 kc./sec. and 100 kc./sec. and the initial permeability is about 385.

19. The method of manufacturing a soft ferromagnetic core material having low magnetic losses in a frequency range between 10 kc./sec. and 100 kc./sec. and increasing magnetic losses above 100 kc./sec. which is particularly suited for cores, inductance coils and the like employed in telephony and telegraphy circuits operating within the said range of frequencies, comprising the steps of heating a first metal oxide and iron oxide forming a first cubic ferrite and a second metal oxide and iron oxide forming a

second cubic ferrite to a temperature between 1000° C. and 1400° C. in an oxygen controlling atmosphere to produce a homogeneous mixed crystal ferrite, and regulating the oxygen content in said mixed crystal ferrite to produce a mixed crystal ferrite material having a coefficient of losses $tg \delta$ which is less than 0.06 in the range of frequencies between about 10 kc./sec. and 100 kc./sec. and greater than 0.06 at a frequency between about 100 kc./sec. and 1000 kc./sec.

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