

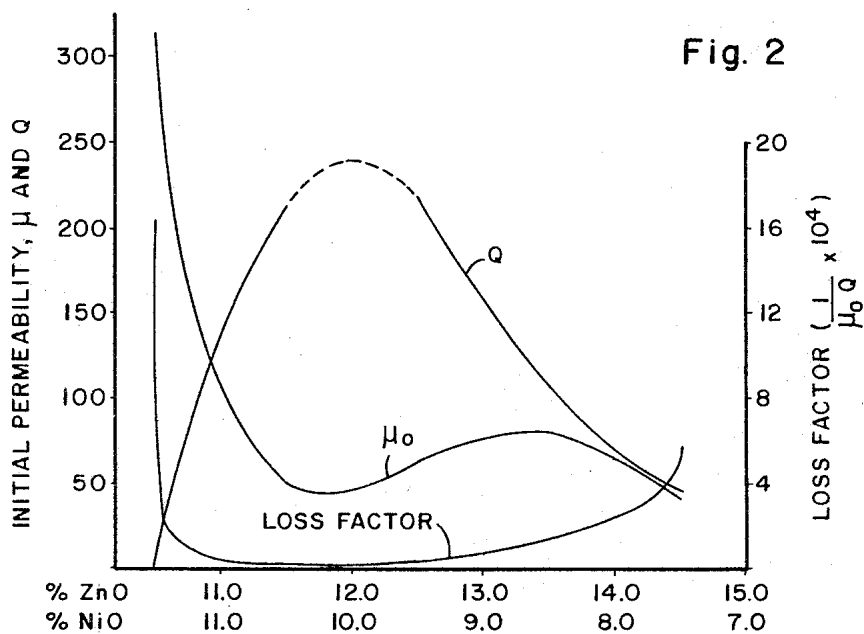
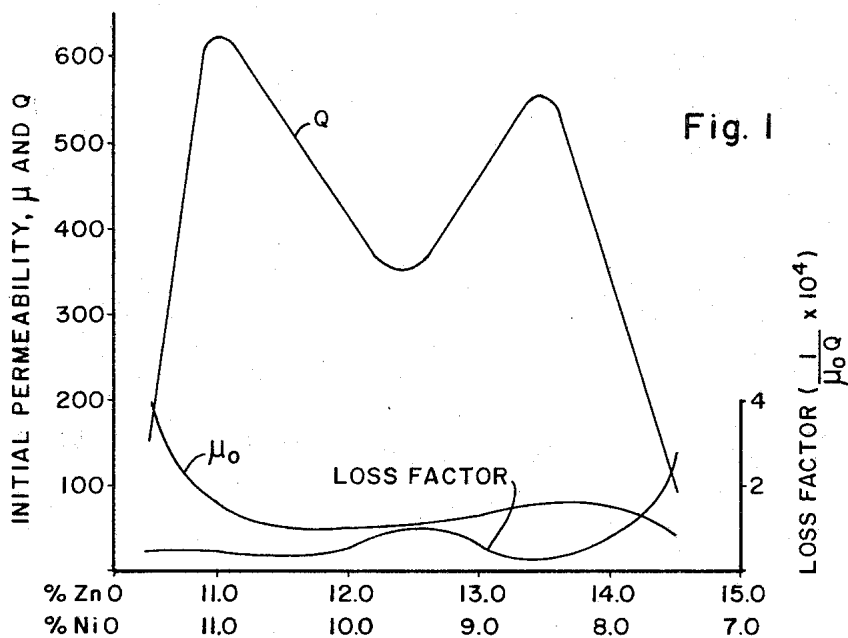
Dec. 27, 1966

W. G. CARLSON ETAL
MAGNETIC CERAMIC MATERIAL

3,294,687

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2 Sheets-Sheet 1



WITNESSES

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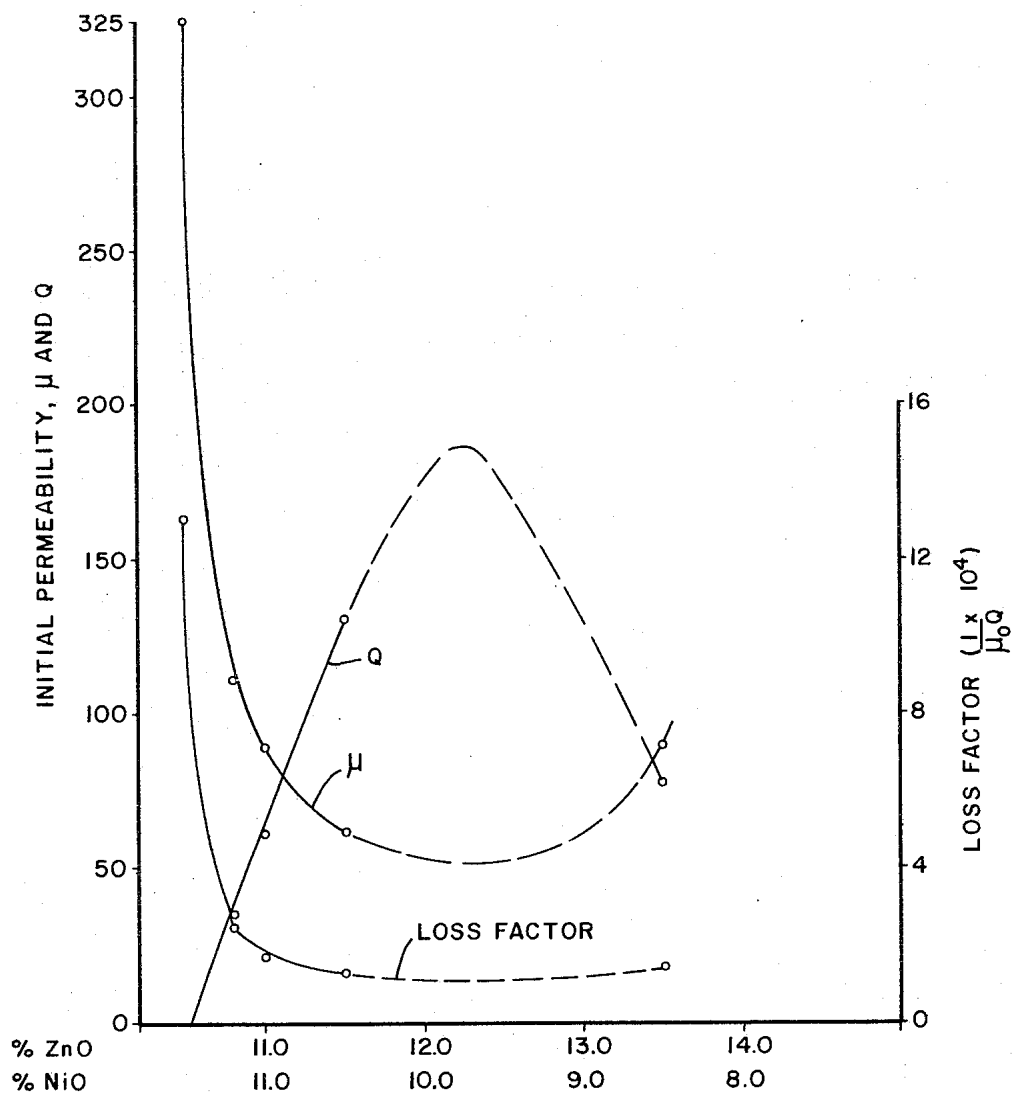


Fig. 3

1

2

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MAGNETIC CERAMIC MATERIAL

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The present invention relates to ferromagnetic oxide bodies and, in particular, to nickel-zinc-manganese sintered ferrites having improved initial permeabilities and improved loss characteristics at relatively high frequencies.

Magnetic bodies consisting essentially of sintered ferrite crystallites, also referred to herein as ferrite bodies or ferromagnetic oxide bodies, are useful in many electronic devices; for example, as cores in antennas, inductors, transformers, and other low loss applications.

In the appraisal of magnetic materials for design applications, and especially for precision inductors, low losses and small changes of permeability and loss values with variations in frequency, flux density, and temperature and over long periods of time are often of more importance than the actual value of permeability. The reciprocal of the $\mu_0 Q$ product of a material has been found to be a useful index of efficiency for design applications. In the product, μ_0 is the initial permeability and Q

$$\left(= 2\pi \frac{fL}{R} \right)$$

represents the reactance of a winding on a core per unit of core loss. The term R is the effective series resistance arising from core loss in the material. The term f is the frequency in cycles per second. It is readily apparent that a high value of $\mu_0 Q$ is desired. A higher value of μ_0 will produce a larger amount of inductance L per turn, which can be used to provide a more efficient coil or a smaller coil of the same efficiency. A high value of μ_0 also reduces leakage flux and improves the shielding when the core material surrounds the winding. In a transformer, a core material with a higher $\mu_0 Q$ results in a proportionally smaller shunt loss across the transformer windings.

In more meaningful terms the initial permeability, μ_0 , is thought of as the electrical energy required to provide a given flux density in a given material or the susceptibility of a given material to flux. The Q factor or quality factor is thought of as the usable flux in a material for a given flux density after deducting inherent losses in the core material and windings along with eddy current losses.

The object of the present invention is to provide a ferromagnetic oxide body having a relatively low loss factor ($1/\mu_0 Q$) at relatively high frequencies.

Another object of the invention is to provide a ferromagnetic oxide ceramic body formed by reacting under heat a compressed mixture of ingredients which on firing will yield a product of by weight 70 to 80% Fe_2O_3 , 10.5% to 14.5% ZnO , 7.5% to 11.5% NiO , up to 1% CoO and 1 to 2% MnO with up to 1% of incidental impurities.

Other objects of the invention will in part, be obvious and will in part, appear hereinafter.

In order to more fully understand the nature and scope of the invention reference should be had to the following detailed description and drawings, in which:

FIGURES 1, 2 and 3 are graphs showing the initial permeability μ_0 , quality factor Q , and loss factor $1/\mu_0 Q$ versus composition at 2, 18 and 30 megacycles respectively.

In accordance with the present invention and in attainment of the foregoing objects, there is provided a ferromagnetic oxide ceramic material comprising the reaction product formed by compressing into pellets or bodies a

mixture of ingredients, which on firing will yield a product comprising essentially by weight of from 70 to 80% Fe_2O_3 , 10.5% to 14.5% ZnO , 7.5% to 11.5% NiO , up to 1% CoO and from 1 to 2% MnO with up to 1% of incidental impurities and heating bodies of the compressed mixture in an oxygen engendering atmosphere, such as, air, at a temperature of from 1100° C. to 1350° C. for the order of two hours. A desirable range of composition is 74 to 78% Fe_2O_3 , 10.5% to 14.5% ZnO , 7.5% to 11.5% NiO , 0.2 to 0.8% CoO and 1.2 to 1.8% MnO . More particularly, there is provided a ferromagnetic oxide ceramic body formed by the reaction products of a mixture of ingredients which on firing will yield a product of by weight 75 to 76% Fe_2O_3 , ZnO and NiO present in the above stated ranges and totalling about 22%, 0.5% CoO and 1.5% MnO with up to 0.5% of incidental impurities.

The ferrites of the invention may be prepared by any means that will ensure homogeneity and a minimum of contamination. Oxides of the metals and, often, their carbonates may be mixed with a fluid medium, usually water, and milled in a steel ball mill. The mill is operated usually between 4 and 20 hours to obtain thorough comminution and intimate mixing of the oxides. The grinding affect in a ball mill on very small particles in the sub-micron size is minor. To minimize introduction of impurities, a rubber lined mill with stainless steel balls is often employed. Although the ball mill is commonly used for the mixing step, other devices such as colloid mills and attritors may be used. Extrusion and pressing additives may be introduced at this point in instances where no presintering is performed and such additives cling to the finely divided oxide particles so that they are not lost when removing the milling medium.

After milling, the homogeneous oxide mixture may be oven dried. To minimize preferential settling of any dense or large particles, the water is rapidly removed. Quite often the slurry is pressure filtered prior to oven drying to expedite the removal.

The dried material may then be presintered, which consists of heat treating the oxide mixture at a temperature somewhat lower than the final firing temperature. After presintering, the oxide mixture is easily comminuted to a particle size that is ceramically workable. Presintering is performed to help control shrinkage in the final shaped members and to influence homogeneity.

Two conventional means of forming the oxide mixture into shapes are die pressing and extruding. With both means, organic additives which are well known in the art are introduced to serve as a binder and particle lubricant. When extruding, a plasticizing agent is also needed. Methyl cellulose, for example, has given fairly satisfactory results as an additive for both pressing and extruding. Polyvinyl alcohol and other organic agents are suitable additives for these purposes.

An effective die design involves a floating die sleeve with top and bottom punches. This permits pressure to be applied from two directions and thereby reduces the tendency to form a packing gradient in the pressed shape. Proper granulation of the die charge also facilitates pacing. Pressures employed may vary from 1 to 10 tons per square inch or even higher depending on the nature of the charge.

After pressing, the shapes should be heated gradually during the low temperature range of the firing cycle to slowly volatilize the organic additive. Rapid evolution of vapors will invariably split the shapes, particularly when porosities are low. Also, under conditions where considerable sintering and shrinkage occur, the rate of heating should be slow.

All ferrites because of the presence of the iron ion, are susceptible to marked changes in oxygen content. The equilibrium between ferric oxide and magnetite shifts

strongly to magnetite above 1370° C. in air. Under reducing conditions the effect is pronounced even at lower temperatures. Therefore, the ferrite bodies are preferably fired between 1100 and 1300° C. so that magnetic formation is not a severe problem particularly where reducing conditions are avoided.

The following example is illustrative of the teachings of the invention.

TABLE I

	A	B	C	D	E	F	G
Fe ₂ O ₃ -----percent..	76	76	76	76	76	76	76
ZnO-----do-----	10.5	10.8	11.0	11.5	12.5	13.5	14.5
NiO-----do-----	11.5	11.2	11.0	10.5	9.6	8.5	7.5
CoO-----do-----	0.5	0.5	0.5	0.5	0.5	0.5	0.5
MnO-----do-----	1.5	1.5	1.5	1.5	1.5	1.5	1.5

A number of ferrite bodies A to G were prepared having the respective compositions listed in Table I. Ferric oxide, cobalt oxide and manganese oxide in the percentages listed were regarded as being nearly at optimum and were therefore kept constant; however, higher and lower percentages of these components from a total of 1% to 2% will give satisfactory results. The nickel oxide and zinc oxide percentages were varied, the total of both of these components being 22% by weight.

The components were weighed out, placed in a steel ball mill with deionized water and a deflocculant, a dispersing agent, and milled with steel balls for eight hours to comminute and blend the several components. The resultant slurry was removed from the ball mill, dried thoroughly and calcined at 1200° C. for one hour to partially react the components to form the ferrite. The calcined material was then crushed to a -20 mesh particle size and again wet ball milled in a steel mill for 20 hours. The milled material was dried completely, screened through a 20 mesh sieve, and mixed dry for 15 minutes with 2% by weight of -100 mesh polyvinyl alcohol. Deionized water then was added slowly to the batch and mixing continued for an additional 20 minutes. After mixing, the batch was stored in a sealed container for 24 hours to give a uniform moisture distribution. After storage, the batch was dried completely and mixed with about 0.2% of a stearate lubricant selling under the tradename Sterotex. The batch is further screened through a 20 mesh sieve onto a 120 mesh sieve. This agglomerate size, within the particle size range of between -20 mesh plus 120 mesh, is used in the pressing operation. The material coarser than 20 mesh and the -120 mesh material can be reprocessed.

In order to facilitate subsequent magnetic testing toroidal samples were pressed; however, other shapes can be fabricated in a similar manner employing the same pressing pressures and sintering temperatures. The toroidal cores were pressed at 15,000 p.s.i. employing a hydraulic press and steel die. The pressed toroids measured 0.690

inch O.D., 0.375 inch I.D. and 0.190 inch in height. The pressed toroids were than sintered in air at 1250° C. for one hour. The heating rate was 180° C. per hour and the cooling rate was determined by the natural cooling rate of the furnace. The toroids underwent a shrinkage of approximately 12% with heat treatment so that the resulting size as sintered was approximately 0.607 inch O.D., 0.330 inch I.D. and 0.167 inch in height. Several toroids from each of the compositions in Table I were measured at frequencies of 2, 18 and 30 megacycles to determine the initial permeability (μ_0), quality factor (Q) and loss factor ($1/\mu_0Q$).

FIGURES 1, 2 and 3 show the effect of varying the nickel oxide and zinc oxide percentage, within a 22% total content, as set forth in Table I, on the initial permeability, Q and the loss factor at 2, 18 and 30 megacycles, respectively. The composition having optimum properties at each frequency may thereby be selected and employed by looking at the curves.

At 18 and 30 megacycles the loss factor increases rapidly when the zinc oxide is reduced from 11% to 10%, and the nickel oxide is increased from 11% to 12%, likewise the loss factor increases rapidly when zinc oxide goes beyond 14% at 2 megacycles or beyond 13% at 18 megacycles.

It is intended that the above description and drawings be interpreted as illustrative and not limiting.

We claim as our invention:

1. A fired ferromagnetic oxide ceramic member consisting essentially of, by weight, 74% to 78% Fe₂O₃, 10.5% to 14.5% ZnO, 7.5% to 11.5% NiO, 0.2% to 0.8% CoO and 1.2% to 1.8% MnO with up to 1% of incidental impurities, the member characterized by low loss factors and high Q factors at high frequencies.

2. A fired ferromagnetic oxide ceramic member consisting essentially of, by weight, 75% to 76% Fe₂O₃, 10.5% to 14.5% ZnO and 7.5% to 11.5% NiO, the ZnO and NiO being present in a combination total of about 22%, about 0.5% CoO and about 1.5% MnO with up to 0.5% of incidental impurities, the member characterized by low loss factors and high Q factors at high frequencies.

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