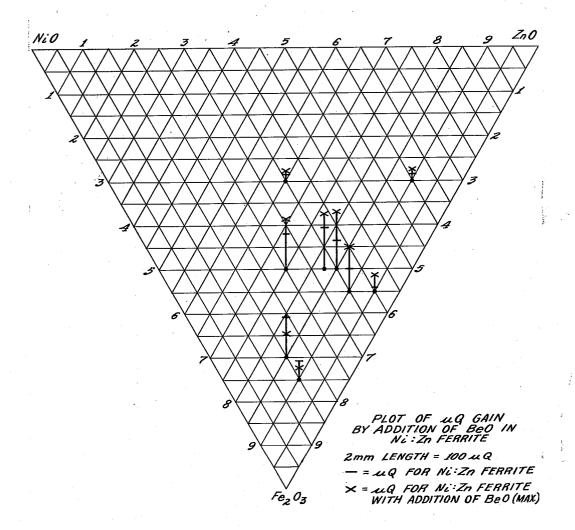
MATERIAL HAVING IMPROVED MAGNETIC PROPERTY
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MATERIAL HAVING IMPROVED MAGNETIC PROPERTY

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9 Claims. (Cl. 252—62.5)

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This invention relates to materials having improved magnetic properties. These materials may have unusually high values of magnetic permeability, or they may have desirably low loss values (high Q factors) at radio frequencies, or a usefully high product of these two values. They may also have desirable properties of magnetostriction and good stability at operating temperatures. They may also have an improved modulus of elasticity and good resistivity.

Wherever Q values are discussed or stated throughout this specification, there is meant a numerical value found by dividing the radio frequency reactance by the resistance of a circuit in which the materials of the present invention are 15 introduced as induction coil core bodies.

More particularly, the present invention relates to materials known as ferrites, made by the general method described in co-pending application of H. W. Leverenz and R. L. Harvey entitled 20 "Improvement in Magnetic Materials," Serial No. 776,292, filed September 26, 1947, now Patent No. 2,565,861.

The present improvement is especially related to the use of beryllium oxide, BeO, either substituted for part of the oxides previously disclosed, in the said application or elsewhere in the prior art, or in addition to the materials therein mentioned

One object of the present invention is to provide compositions having improved magnetic properties.

Another object of the invention is to provide materials having controllable and unusually high values of magnetic permeability.

Another object of the invention is to provide materials having low loss values (high Q factors).

Another object of the invention is to provide materials having improved magnetic properties and also high Curie temperatures.

Another object of the invention is to provide materials having a desirably wide range of magnetostriction.

Another object of the invention is to provide materials having improved magnetic properties along with a desirable modulus of elasticity.

Another object is to provide improved compositions of magnetically permeable ferrites which include beryllium oxide as an essential ingredient.

Still another object is to provide materials having desirable values of resistivity.

These and other objects will be more readily apparent and the invention will be better understood with reference to the following specification, including the drawing, of which the single figure is a graphical representation of the improvement in the product μQ obtained by addition of BeO in a Ni-Zn ferrite.

described in the previously mentioned co-pending application, Serial No. 776,292, the general method of preparation is the same as illustrated therein. Briefly, this method includes the intimate mixing of ferric oxied and the various other oxides involved, each in a fine state of subdivision, heating, preferably, a compressed body made of these oxides within a range of temperatures of about 900° C. to about 1500° C. for about 1 minute to about 5 hours in an oxidizing atmosphere, and then cooling. Longer heating times are required for larger bodies. Optimum conditions of time and temperature of heating will vary with each specific composition possible within the systems of compositions which are a part of the present invention and, with any specific composition, the optimum heating time decreases as the temperature of heating increases. In general, however, some improvement in results is obtained by selecting widely varying times and temperatures within the ranges specified, even though these improvements will not be the maximum obtainable when a more careful choice is

Although the compositions, which are a part

of the present invention, are different from those

about 1 hour is used, and a heating temperature of the order of 1200° C. has proved satisfactory, if not always best, with all of the compositions. The reaction product which is fromed in all instances is a composite, homogeneous, crystalline body winch does not have the same stoichiometric proportion of oxygen to metals as is present in the mixture before heating, but the exact proportions of the elements present in the products is not known and cannot easily be determined.

made. Good results can be expected with almost

any of the compositions, if a heating time of

The oxidizing atmosphere in which the heating of the reaction mixture takes place may be supplied by passing a stream of oxygen through the reaction chamber. Less desirably, air may be used in place of oxygen. It is also possible to form products having improved magnetic permeability and low loss by heating the oxides in a neutral atmosphere, such as one comprising helium or nitrogen, but the improvement is not as great as when an oxidizing atmosphere is used. In contrast to the use of either an oxidizing or a neutral atmosphere, the presence of a reducing atmosphere, such as supplied by carbon monoxide or hydrogen, in the reaction chamber, is distinctly detrimental, if materials having high permeability are desired. Therefore, it may be said that the heating should take place in a nonreducing atmosphere, preferably at atmospheric or higher pressures.

Instead of starting with the oxides themselves, it is possible to start wwith a mixture of mate-

rials in other than the oxide form in which they are present in the reaction product, provided these starting materials will be changed to the specified oxide upon heating to the temperatures, times and atmospheric conditions stipulated. 5 For example, instead of starting with a mixture containing Fe2O3, the iron may be in the form of ferrous oxide or magnetite, providing that heating takes place in an oxidizing atmosphere, and as the hydroxide, carbonate, etc., if either 10 an oxidizing or a neutral atmosphere is present. The other ingredients may also be present in forms such as hydroxides, acetates or carbonates or either lower or higher oxide forms than are present in the final product, since these will re- 15 relates more specifically to the use of beryllium vert to the desired oxide form when heated strongly in the preferred oxidizing atmosphere and, in some cases, even in a neutral atmosphere. Certain of the present compositions also utilize oxides of manganese. This is preferably present 20 as MnO2 in the initial mixture but changes to MnO in the reaction product. This change occurs even though an oxidizing atmosphere be used, providing the oxidization properties of the atmosphere are not strong enough to prevent the 25 reduction of the higher valent oxide to the lower divalent form. Likewise, other oxides of manganese can be used in the starting mixture since these will revert to MnO when heated in the oxidizing atmosphere. Similarly, it is possible to use 30 oxide of copper or nickel since there also will change to the desired form when heated under the oxidizing conditions preferred.

Because of the brittleness of the reaction products, compressed bodies of the reactants may be formed at pressures of, say, 20,000 pounds per square inch and these compressed bodies are treated at elevated temperatures in order to impart the desired properties of increased permeability, low loss, etc., desired in the end product. These compressed bodies may also be formed by extrusion molding processes at much lower pressures. The fact that shaped bodies of the compressed material can be formed without the presence of a permanent binder adds much to their improvement over materials previously known, such as powdered iron, which requires the use of a permanent binder, since the binder introduces a multitude of gaps of considerable size between the particles, and the gaps, themselves, are re- 50 sponsible for much of the lower permeability hitherto present in materials of this nature. When the material is made in the form of compressed bodies, the pressure of forming may vary widely. Pressures of 2,000 pounds per square inch 55 have been found to produce the same improved results as those ten times as great and extrusion molding techniques have been used with excellent results. For example, the ingredient oxides may and extrusion molded and heat treated. In general, it may be said that the pressure of forming should be sufficient to form a closely coherent body and the pressures used should simply be those necessary to obtain this result.

After the heat treatment has been accomplished as described, the reaction product is preferably, although not necessarily, cooled very rapidly, as by subjecting it to a blast of air or quenching in water. Rapid cooling generally increases the permeability and low loss qualities above those values obtained when slow cooling is used.

The reaction mixture may be compressed into

the reaction chamber. The danger of warping of the compressed bodies, when heated, may be lessened by mixing a small percentage of a lubricant with powdered oxides before compression. The preferred lubricants are those which are completely burned off during the heat treatment process. Suitable examples are stearic acid and microcrystalline waxes, such as Carbowax.

Other improvements in molding technique are described in the previously mentioned co-pending application, such as releasing pressure uniformly from the mold piston after compressing the oxide mixture into the desired shape.

As previously stated, the present invention oxide in ferrite bodies, prepared as described. In a wide range of compositions, it has been found that the addition of beryllium oxide produces desirable improvements not obtainable when mixtures of oxides, not including beryllium oxide, are used.

One of the new systems, according to the present invention, is a quaternary system comprising BeO, MnO, ZnO and Fe₂O₃. In Table I below are given examples of various compositions of these four ingredients expressed in mole parts. First, there is given a composition with the beryllium oxide omitted and this is followed by compositions in which various small proportions of beryllium oxide have been substituted for part of the zinc oxide, with the proportions of the other two ingredients remaining constant for purposes of comparison. In each case, in all of the tables which follow, the values of several magnetic properties are given: i. e., effective permeability µeff., the reciprocal of the loss value Q, the product μQ , and, in a few instances, the Curie temperature. Since the Curie temperature is a temperature at which a marked change of properties occurs, probably due to crystalline reorganization, it is desirable to have the Curie temperature of a composition as far above operating temperatures as is convenient since, if the Curie temperature falls within the range of operating temperatures, the magnetic properties may vary quite widely when the material is in use. The values in Table I were obtained by crystallizing the materials at 1200° in an oxygen atmosphere.

Table 1

J.		*****		****				
	BeO	ZnO	MnO2	Fe ₂ O ₃	μett.	Q	μQ	Curie Temp.
5.	0 0.025 0.075 0.125	0. 25 0. 225 0. 175 0. 125	0. 25 0. 25 0. 25 0. 25	0.5 0.5 0.5 0.5	15. 9 16. 0 14. 6 12. 8	40 55 105 117	636 880 1533 1498	° C. 115 124 200

In the above system, is has been found that the be pre-heated, mixed with a temporary binder 60 physical nature (e.g., density, particle size, degree of aggregation) of the manganese dioxide influences the maximum μ and Q and the rate of attainment of the maximum μ and Q. Dense particles of moderate size, such as 1 to 10 microns, are preferable to very small particles (e.g., less than 0.1 micron). The degree of purity of the MnO2 and other ingredients is also an important factor, high purity being desirable. During the crystallization process, brought about by the heat 70 treatment, the MnO2 decomposes to afford MnO.

BeO was also substituted for part of the MnO2 in preparing materials falling within the BeO:ZnO:MnO:Fe2O3 system with results shown by the examples listed in Table IA. It will be seen any desired shape before heating takes place in 75 that here, also, the use of BeO resulted in com-

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positions having improved magnetic properties. Crystallization was carried out at 1200° C. in oxygen.

Table IA

BeO	ZnO	MnO ₂	Fe ₂ O ₃	μeti.	Q	μQ
0	0. 25	0. 25	0. 5	15. 5	36	558
0.02	0. 25	0. 23	0. 5	15. 6	45	702
0.10	0. 25	0. 15	0. 5	14. 9	86	1281
0.20	0. 25	0. 05	0. 5	8. 8	87	766

Keeping the ZnO and MnO₂ content constant, BeO was also substituted for part of the Fe₂O₃ in preparing materials within the system

BeO:ZnO:MnO2.Fe2O3

with results illustrated in Table IB. Crystallization was carried out at 1200° C. in oxygen.

Table IB

-	BeO ZnO		MnO ₂	Fe ₂ O ₃	μ _{eff} .	Q	μQ	
	0. 02 0. 05	0.75 0.25	0. 25 0. 25	0. 48 0. 45	15. 8 15. 2	37 27	585 410	

Another modification in the method of preparing crystalline materials within the system

BeO:ZnO:MnO2:Fe2O3

was carried out by adding BeO with molecular proportions of all of the other ingredients remaining constant. Crystallization was carried out at 1200° C. in oxygen. Data are shown in Table IC.

Table IC

					·	<u></u>	1
BeO	ZnO	MnO ₂	Fe ₂ O ₃	μeff.	Q	μQ	
0. 01 0. 04 0. 06 0. 20	0. 25 0. 25 0. 25 0. 25 0. 25	0. 25 0. 25 0. 25 0. 25 0. 25	0. 5 0. 5 0. 5 0. 5	15. 6 15. 5 15. 7 14. 8	45 44 46 65	702 682 722 962	

In Table II below, values are given for a crystalline system comprising

BeO:ZnO:NiO:Fe2O3

The values in this table were obtained by crystallization of the materials at 1400° C. in an oxygen atmosphere.

Table II

							100
BeO	ZnO	NiO	Fe ₂ O ₃	μeff.	Q	μQ	Curie Temp.
0 0.02 0.125 0.3 0 0.05 0.125 0.125 0.125 0.125 0.02 0.1 0.05 0.05 0.1 0 0.1 0 0.05 0.1 0 0.05	0. 35 0. 33 0. 225 0. 15 0. 05 0. 22 0. 125 0. 325 0. 325 0. 35 0. 15 0. 10 0.	0. 15 0. 15 0. 15 0. 15 0. 25 0. 25 0. 27 0. 175 0. 175 0. 1 0. 1 0. 05 0. 035 0. 035 0. 035 0. 035 0. 1 0. 1 0. 1 0. 1 0. 1 0. 1 0. 1 0. 1	0.55 0.55 0.55 0.55 0.55 0.55 0.55 0.55	15. 2 14. 4 8. 4 8. 4 3 12. 5 12. 2 14. 9 14. 3 13. 2 13. 2 4. 0 14. 0 15. 1 15. 2 16. 3 17. 3 18. 4 18. 4 18. 4 18. 4 18. 4 18. 4 18. 5 18. 6 18. 6 1	39 722 82 76 102 54 85 95 59 77 86 30 31 31 38 48 18 30 111 77 83 75	593 1090 1180 638 337 675 1037 874 879 1101 1084 399 409 409 152 197 131 180 810 810 850 813	° C. 80 130 1276

In the above table, it will be noted that data are given for compositions in which various percentages of ferric oxide are used and also in which the nickel oxide and zinc oxide are varied widely.

In Table III, values are given for a system comprising BeO:ZnO:MgO:Fe₂O₃. These values were obtained by crystallization at 1300° C. in an oxygen atmosphere.

Table III

	BeO	ZnO	MgO	Fe ₂ O ₃	μett.	Q	μQ	Curie Temp.
15								° с.
	0 0.02 0.1	0. 25 0. 25 0. 15	0. 25 0. 25 0. 25	0. 5 0. 5 0. 5	15. 1 13. 2 10. 3	44 57 65	664 752 670	115 356
	0.125 0.2	0. 125 0. 05	0. 25 0. 25	0. 5 0. 5	9. 2 4. 4 1. 2	60 75 27	552 330 32	
20	0 0.05 0	0.35 0.3 0.3	0. 15 0. 15 0. 3	0.5 0.5 0.4	8.3 10.0	30 23	249 230	==
	0.02 0.05	0. 28 0. 25 0. 35	0.3 0.3 0.35	0.4 0.4 0.3	11.8 9.5 5.0	18 37 8	212 352 40	
	0 0.05 0	0.3 0.2	0.35 0.2	0.3 0.6	6.0 9.2	22 70	132 644	
25	0. 02 0. 05 0	.0.18 0.15 0.225	0. 2 0. 2 0. 225	0.6 0.6 0.55	9. 7 8. 2 12. 1	65 75 65	631 615 786. 5	
	0.05	0. 225 0. 175 0. 3	0. 225 0. 05	0.55 0.65	10.5 10.9	75 50	788 545	
	0.02	0.28	0.05	0.65	8.6	95	817	1

Table IV contains values obtained for a system comprising BeO:ZnO:CuO:Fe₂O₃. The values were measured for materials crystallized at 1200° C. in an oxygen atmosphere.

Table IV

6.4							4 4 1
BeO	ZnO	CuO	Fe ₂ O ₃	μeff.	Q	μQ	Curie Temp.
0 0.02 0.07 0.1 0.15 0 0.05 0 0.02 0.05 0 0.05	0.3 0.28 0.23 0.2 0.15 0.35 0.3 0.4 0.35 0.35 0.35 0.35 0.35	0. 2 0. 2 0. 2 0. 2 0. 15 0. 15 0. 1 0. 1 0. 1 0. 2 0. 2	0.55 0.55 0.55 0.55 0.55 0.55 0.55 0.45 0.4	14. 2 14. 2 15. 2 14. 4 12. 5 1. 04 4. 0 1. 2 1. 02 1. 06 1. 8 15. 7 14. 9	20 30 59 73 72 53 20 29 35 57 50 20 42 60	284 426 897 1, 051 900 55 80 35 42 58 53 36 659 894	° C. 40 130

Table V contains values for a few examples of compositions within the system

BeO:CdO:CuO:Fe₂O₃

Crystallization was at 1200° C. in an oxygen atmosphere.

Table V

В	eO	CdO CuO Fe ₂ O ₃		Fe ₂ O ₃	μeff.	μeff. Q		
0.	02 05 10	0.30 0.28 0.25 0.15	0. 20 0. 20 0. 20 0. 20	0. 50 0. 50 0. 50 0. 50	13. 6 14. 0 14. 2 12. 0	20 25 35 71	272 350 497 852	

It will be noted that for small additions of BeO to replace some of the CdO, the permeability rises and so does the Q value. When the amount of BeO is increased, the permeability drops but the Q value rises so rapidly that the μQ product is greatly improved.

Table VI contains examples within the system BeO:CdO:MnO:Fe₂O₃. Crystallization was at 1200° C. in an oxygen atmosphere.

7 Table VI

1.0							
BeO	CdO	MnO^2	Fe ₂ O ₃	μeff.	Q	μQ	
0. 10 0. 05 0. 10	0. 25 0. 20 0. 15	0, 25 0, 25 0, 25	0. 50 0. 50 0. 50	15. 6 13. 7 10. 6	26 60 65	406 822 689	

In these compositions, marked improvement in **Q** value is evidenced when BeO is added to the basic ternary system.

Table VII shows the effects of adding BeO to the system MnO:ZnO:CdO:Fe₂O₃ to form a pentanary system including BeO. The Q value of the materials is raised considerably. Crystallization was at 1200° C, in an oxygen atmosphere.

Table VII

BeO	ZnO	CdO	CdO MnO2		μeff.	Q	μQ	
0 0. 05	0. 125 0. 10	0. 125 0. 10	0. 25 0. 25	0. 5 0. 5	15. 4 14. 3	29 72	447 1030	

In all of the above systems, the beryllium oxide was added to the mixture before the mixture was compressed and crystallization carried out. In general, it has been found that the addition of about 0.2 to about 30 mole percent of co-crystallized BeO improves the product μQ ("figure of merit") of the ferrite solid, and somewhat higher values (e. g., up to about 50 mole-percent BeO) may be used to obtain improved Q values, although the product μQ may not be improved when the highest percentages of BeO are used. When a composition of the three ingredients, without beryllium oxide, is selected, which is near the optimum for maximum μ , it has been found that the optimum proportion of co-crystallized BeO is about 3 to about 15 mole percent. When the ternary-system composition is high in iron oxide, the addition of BeO may lower the product μQ ; hence, the improved materials should preferably comprise no more than about 70 mole-percent of iron oxide. Furthermore, compositions which have less than about 30 molepercent of Fe₂O₃ have poor characteristics which are generally not improved sufficiently by the incorporation of BeO to be useful. Perhaps the greatest improvements, on incorporating BeO, occur in those compositions having Curie temperatures near room temperature. As shown in the above tables, the incorporation of BeO beneficially raises the Curie temperature, as well as the product μ Q. This useful effect of BeO extends the range of useful compositions which are usable at a given temperature.

The improvement in the product μQ brought about by the inclusion of BeO in the ferrite systems is illustrated graphically in the figure. The figure shows the extent of improvement that it is possible to obtain by adding an optimum amount of BeO to ferrites of the system NiO:ZnO:Fe2O3. The examples given are those in which an amount of BeO is added such that the maximum increase in value of μQ is obtained. It will be noted that a composition made up of 20 a mixture of 50 mole percent Fe₂O₃, 15 mole percent NiO, and 35 mole percent ZnO can have its μQ product increased by about 530 units when an optimum amount of BeO is included in the composition before crystallization. It will be noted further that, when the compositions contain about 30 mole percent Fe₂O₃, the maximum possible improvement is very small, while, for compositions containing above about 70 mole percent Fe₂O₃, the addition of BeO may lower the μ Q product, no matter what amount is added. The figure also illustrates the relative improvement possible to bring about by adding BeO since the increase in value sometimes amounts to several hundred percent.

Although the greatest improvement in magnetic properties has been found when BeO is included in a quaternary system type ferrite, it has also been found that compositions having usefully improved magnetic properties can be obtained by utilizing varying percentages of BeO in a ternary system. Data illustrating examples of various ternary system ferrites, including BeO, are given in Table VIII. It will be noted that some significant improvement is obtained in almost all instances by the addition of BeO although the percentage of improvement is not as great, in general, as when the BeO is added or substituted to form a quaternary system.

Table VIII

					4	<u> </u>			
Fe ₂ O ₂	MnO2	ZnO	MgO	NiO	CuO:	BeO	μeff.	Q	μQ
0. 45 0. 45 0. 45	0. 55 0. 53 0. 45					0. 02 0. 10	11.9 11.8 11.2	85 20 53	1012 236 594
0.5 0.5	0. 5 0. 45					0.05	13. 0 11. 6	35 75	405 870
0. 55 0. 55 0. 55	0. 45 0. 43 0. 40					0. 02 0. 05	12.1 9.1 7.7	5 81 66	61 737 508
0.45 0.45		: 		0. 55 0. 50		0.05	3.5 4.0	61 115	214 460
0. 50 0. 50 0. 50				0.50 0.45 0.40		0. 05 0. 10	4.3 4.4 4.0	75 90 106	323 396 424
0.60 0.60				0.40 0.30		0.10	6. 1 3. 5	94 127	573 445
0.70				0. 25		0.05	3.6	115	414
0.40 0.40			0.60 0.45			0.15	5. 3 6. 5	43 63	228 410
0. 50 0. 50			0.50 0.40			0.10	6. 7 4. 2	55 75	369 315
0.60 0.60			0. 40 0. 30			0.10	3. 5 2. 8	90 100	315 280
0.45 0.45					0. 55 0. 45	0.10	7.4 6.4	25 40	185 256

Table VIII—Continued

•			5	rable 1	viii—	Contin	ued		19 19		
	Fe ₂ O ₃	MnO ₂	ZnO	MgO	Nio	CuO	BeO	μeff.	Q	μQ	
	0.50					0.40	0.10	6.4	43	275	e profession engineering. Annother the street of the
	0.60 0.60					0.40 0.30	0.10	10.8 10.4	48 80	518 832	
	0.65					0.30	0.05	10.8	67	734	attivities of projection and
	0. 5 0. 6 0. 65		0. 25 0. 35 0. 30				0. 25 0. 05 0. 05	5. 2 10. 9 9. 4	60 27 42	312 294 395	jedyčeden y s ačeje stanie Postava sakodenje se Postava sakodenje se

All of the above values were obtained by adding BeO to the various oxide mixtures and then 15 forming compressed bodies and heating in an oxidizing atmosphere. The heating temperature was 1200° C. except that those compositions containing NiO were heated at 1400° C., those containing MgO were heated at 1300° C. and those 20 containing ZnO were heated at 1400° C.

From the data given in the above table, it can be seen that, in some cases, the addition of BeO to a binary system ferrite does not improve the magnetic properties as much as by adding it to 25 a ternary system and in a few cases a slight decrease is noted. However, the proper standard of comparison is a similar body made of powdered iron which was formerly the best material known for high magnetic permeability. In all cases, the 30 materials composed of the ternary system ferrites, including BeO, evidenced improved properties when compared to similar bodies made of powdered iron compressed with a binder. Moreover, for special uses, ternary system products, 35 including BeO, have been found best since the Q values are usually high and the μ Q product is also sufficiently high to be useful.

Of all of the above systems which form a part of the present invention, a quaternary system is 40 preferred in which BeO and ZnO are both present. These compositions have proved best as to stability, permeability and Q value. The addition of BeO, and to a lesser extent MgO, has been found to be the only factor which can be added to the ferrite systems in order to improve 45 the product μQ without any loss (or undue loss) of u.

Addition of many other oxides has been tried. The addition of CaO, BaO, TiO2, and ZrO2 usually lowers both effective permeability, μ , and μQ . 50 Addition of any of ThO2, Sb2O3, Al2O3, SiO2, SnO, B₂O₃ or Cr₂O₃ sometimes gives compositions having moderately improved μQ but the effective permeability is usually undesirably lowered.

Although it it is not desired to be restricted to 55 the following theory, it is believed that the beneficial effects of adding BeO (or MgO) to the ferrite compositions may be related to the higher polarizing power of the Be++ (or Mg++) ions and the higher melting points of the BeO (or 60 MgO). The polarizing power, for example, is proportional to q/r, where q is the valence charge and r the ionic radius. Be++ exhibits an outstandingly high ratio of q/r.

Another interesting and important property of 65 compressed bodies made up of compositions falling within the present invention is that of magnetostriction. Magnetostriction is evidenced by the ability of a body to lengthen in one dimension and contract in another direction when sub- 70 jected to a magnetic field. Measurements were made on compressed cores of the materials inserted within a hollow tube 11/2 inches long and having 1/4 inch outside diameter. The cores were rectangular in cross section and had a length of 75 therein. These have been chosen for purposes

21/2 inches. The tube was wound with a solenoid of 990 turns of No. 30 copper wire. Measurements were taken using two different field strengths, namely, 49.0 oersteds (M1) and 150.5 oersteds (M2). The former was obtained by applying a battery current of 0.15 ampere at 1.5 volts and the later field was obtained by applying a battery current of 0.46 ampere at 6 volts to the solenoid.

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Some of the core bodies exhibited only positive magnetostriction, as evidenced by increase in length, and others only negative, while some exhibited positive at one field strength and negative at another. In the following table (Table IX) some examples of typical results are given, the positive values, that is lengthening, being designated -P- and negative values, that is shortening, being designated —N—. All values are relative, a bar of nickel of similar dimensions being taken as a standard and assigned the value 4N when subjected to a field of 49 oersteds. A system of levers was used to magnify the amount of dimensional change and the amount of the change was measured by observing the movement of a knife edge connected to the lever system. The units expressed in the table are simply taken in terms of the gradations of the scale of the observing scope.

Table IX

	100 0000		1900			1 2 2 2	: 2	<u> </u>	
BeO	ZnO	MnO2	F	2O3	49 oersteds M ₁		150.5 oersteds M ₂		
0. 025 0. 075 0. 125	0. 225 0. 175 0. 125	0. 25 0. 25 0. 25		0. 5 0. 5 0. 5		0.7N 1.7P 2.0P		1.3N 1.0N 0.4N	
BeO	ZnO	Ni	0	Fe	O ₃	M ₁		M_2	
0 0. 02 0. 125 0. 2 0. 3	0. 35 0. 33 0. 225 0. 15 0. 05	0.1	15 15	0. 0. 0.	5 5 5	0.5N 1.3N 1.8N 1.3P 0.3P		0.8N 1.6N 1.8N 0.7P 0.6P	
BeO	ZnO	Mg	0	Fe ₂ O ₃		M ₁		M ₂	
0 0.02 0.1 0.2 0.1 0.15 0.3	0. 25 0. 23 0. 15 0. 05 0. 2 0. 15 0	0. : 0. : 0. : 0. : 0. : 0. :	25 25 25 25 2	0. 5 0. 5 0. 5 0. 5 0. 5 0. 5 0. 5		0.5N 1.5P 0.75P 1.0P 0.7N 0.6N 0.25P		0. 7N 0. 8P 1. 8P 0. 8N 1. 0N 0. 1P	

From the figures in Table IX, it can be seen that, if a body having a certain magnetostriction is desired, it is simply necessary to select the proper composition and prepare a crystalline ferrite body in the manner described herein.

The above examples contained in the several tables are not to be construed as limiting the invention to the specific compositions listed of illustration only and have been selected from a larger mass of data. The approximate limits of practical operation have been pointed out elsewhere above and it is these limits as well as those found in the appended claims which it is desired to encompass within the present invention.

In general, the present invention constitutes the discovery that the addition of beryllium oxide to crystalline ferrite materials containing at least 10 one other oxide of a certain group of oxides results in the formation of materials having improved magnetic properties, especially if compared with the best previously known materials formerly used for the same purposes, such as 15 powdered iron. The group of other oxides from which one or more may be selected for inclusion in the compositions is MnO, ZnO, CuO, CdO, NiO and MgO. These may be present in the form of equivalent compounds in the original mixture 20 but must be readily changed to the above oxide form during the heating operation required to bring about the crystal formation.

Especially good results are obtained by selecting two of the above group of oxides together 25 with BeO and experiments indicate that three or more may also be used although, apparently, no additional advantage is gained by so doing.

The improvement in results obtained through use of the present compositions may be in any one or more of several categories; hence, whether or not a certain material constitutes an improved product compared with prior art materials cannot be judged by any one test. Certain of the compositions exhibit exceptionally high values of magnetic permeability, others have exceptionally low loss factor (high Q), some have a desirably high product of these two values, others have desirable magnetostrictive properties and still others have great stability although their permeability or Q values may not be as high as any one of a number of other possible compositions. In some cases, a composition may have exceptionally high Curie temperature and thus be useful for certain applications.

We claim as our invention:

- 1. A composition of matter comprising the reaction product produced by heating together in a non-reducing atmosphere at temperatures of 900° C. to 1500° C. for 1 minute to 5 hours an intimate mixture of from 30 to 70 mole percent Fe_2O_3 , 0.2 to 50 mole percent BeO, and the remainder comprising at least 20 mole percent one other oxide from the class consisting of the oxides of manganese, zinc, magnesium, nickel, cadmium and copper.
- 2. A composition, according to claim 1, in which the amount of BeO is from about 0.2 to about 30 mole percent.
- 3. A composition, according to claim 1, in $_{60}$ which the amount of BeO is about 3 to about 15 mole percent.
- 4. A composition of matter comprising the reaction product produced by heating together in a non-reducing atmosphere at temperatures of 900° C. to 1500° C. for 1 minute to 5 hours an intimate mixture of about 30 to 70 mole percent Fe₂O₃, about 0.2 to about 50 mole percent BeO, and the remainder comprising at least 20 mole percent of one oxide from the class consisting of MnO₂, ZnO, MgO, NiO, CuO and CdO.
- 5. A composition of matter comprising the reaction product produced by heating together in a non-reducing atmosphere at temperatures of 900° C. to 1500° C. for 1 minute to 5 hours an

intimate mixture of about 30 to about 70 mole percent Fe₂O₃, about 0.2 to 50 mole percent BeO, and the remainder comprising at least 20 mole percent of two oxides of different metals from the class consisting of oxides of manganese, zinc, magnesium, nickel, copper and cadmium.

6. A composition of matter comprising the reaction product produced by heating together in a non-reducing atmosphere for 1 minute to 5 hours at temperatures of 900° C. to 1500° C. an intimate mixture of about 30 to about 70 mole percent Fe₂O₃, about 0.2 to about 50 mole percent BeO, and the remainder comprising at least 20 mole percent of ZnO and at least one oxide from the class consisting of the oxides of manganese, magnesium, nickel, copper and cadmium.

7. An article of manufacture characterized by having improved magnetic properties comprising a compressed body of material of predetermined shape, said material comprising a reaction product produced by heating together in a non-reducing atmosphere at a temperature of 900° C. to 1500° C. for 1 minute to 5 hours an intimate mixture of 30 to 70 mole percent Fe₂O₃, 0.2 to 50 mole percent BeO and at least 20 mole percent of an oxide from the class consisting of the oxides of manganese, zinc, magnesium, nickel, copper and cadmium.

8. An article of manufacture characterized by having improved magnetic properties comprising a compressed body of material of predetermined shape, said material comprising a reaction product produced by heating together in a non-reducing atmosphere at a temperature of 900° C.
 35 to 1500° C. for 1 minute to 5 hours an intimate mixture of 30 to 70 mole percent Fe₂O₃, 0.2 to 50 mole percent BeO, the remainder comprising at least 29 mole percent ZnO and at least one other oxide from the class consisting of the 40 oxides of manganese, magnesium, nickel, copper and cadmium.

9. A method of forming a core body characterized by having improved magnetic properties comprising preparing an intimate mixture of 30 to 70 mole percent Fe₂O₃, 0.2 to 50 mole percent BeO and at least 20 mole percent of an oxide from the class consisting of MnO₂, ZnO, MgO, NiO, CuO and CdO, compressing said mixture to form a coherent molded body of predetermined shape, subjecting said molded body to a temperature of 900° C. to 1500° C. in a non-reducing atmosphere for 1 minute to 5 hours to form a crystalline substance having a definite chemical composition, and cooling said crystalline substance.

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REFERENCES CITED

The following references are of record in the file of this patent:

UNITED STATES PATENTS

Number Name Date 1,946,964 Cobb Feb. 13, 1934 65 1,976,230 Kato Oct. 9, 1934 OTHER REFERENCES

Snoek: "Magnetic and Electrical Properties of the Binary Systems Mo.Fe₂O₃," published in

70 Physica, III, No. 6, June, 1936, pages 481-482. Mellor: "Comprehensive Treatise on Inorganic and Theoretical Chemistry," vol. 14, 1934, page 914.

a non-reducing atmosphere at temperatures of Kawai: Journal of the Society of Chemical In-900° C. to 1500° C. for 1 minute to 5 hours an 75 dustry, Japan, vol. 37, No. 4, 1934, page 174B.