THE EFFECTS OF SOME ADDITIVES AND IMPURITIES ON THE MAGNETIC PROPERTIES OF MANGANESE—ZINC FERRITES

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Summary

Additives have come into the limelight of ferrite-development and complex additives, containing several components, are to be successfully used to improve manganese—zinc ferrites. Some impurities are regarded as useful additive-components. In case of the M2 type manganese—zinc ferrite material (manufactured by HAGY) losses have substantially been reduced as well as temperature and time stability bettered by using a complex additive containing Ta_2O_5, CaO and SiO_2 (the two latter oxides are impurities in ferrite raw materials).

Introduction

Ever since their advent in the early nineteen-twenties, soft magnetic ferrites have fully developed and have come to replace ferromagnetic metals in telecommunication technology. Not only have they eliminated the drawbacks of the latter, i.e. losses due to eddy-currents, noisiness in transmission and distortion of weak telecommunication signals but they have also acquired valuable properties as, e.g. high initial permeability, sensitivity to receive and to discriminate various pulses carrying all kinds of information and capacity to reproduce magnetic and electrical events, having activated them before.

As a matter of fact, mixed soft ferrites have marked a new era for magnetic materials and exceedingly good results have been obtained with manganese—zinc ferrites as well. All over the world, thousands of ferrite-researchers have taken part in extensive investigations and have contributed to their success. As a result, these ferromagnetics are nowadays widely used in a number of telecommunication fields.

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Scrutinizing the ways and means leading to the high perfection of soft ferrites it is obvious that the ferrite structure has a dominant priority in this respect. First of all magnetic ferrites are dielectrics and have, consequently, high electrical resistivities. This accounts for the fact that ferrites are to a high degree free from eddy-current losses. The base of the polycrystalline ferrite structure is a spinel-lattice consisting of oxygen-ions. In the sites between the latter various metal-ions are introduced during the ferritizing process and classical ferrite-research has already cleared up how the magnetic moments of these ions i.e. their sub-lattices are summed up. That is why the general model of ferrite-structure has become well-known to experts and also methods and techniques have been found to influence or modify it. At times, it has proved to be possible to produce ferrites having anticipated magnetic and other properties and no doubt this is the common aim of all research-workers interested in ferrite-development. However, because of international rivalry it is by no means easy to improve or perfect present-day advanced ferrites. Under such conditions most competitive experts stand on the alert to take immediate advantage of the slightest advances they have come to know of. In addition, there are phenomena in ferrite-technology that must be cleared up first. Some of the experts liken ferrite-making to cookery itself, saying that a first-rate cook can make a delicious dish of given ingredients while others cannot and the same holds for ferrite-manufacturing, too.

Manganese—zinc ferrites are most interesting from this point of view since they react to technological measures rather promptly.

Present-Day Development Trends of Manganese—Zinc Ferrites

Among soft magnetic materials, manganese—zinc ferrites represent a product-family suiting most telecommunication technology applications. Owing to their favourable properties (relatively high resistivity, almost complete lack of eddy-currents, good temperature stability, etc.), these ferromagnetics are of great importance not only for up-to-date telecommunication purposes, but also for entertainment electronics. In addition, the raw materials they are made from are cheap and their manufacturing costs are not inhibitive. Compared to other soft ferrites, they excel in having a high initial permeability, low hysteresis losses and a relative temperature — independence of permeability. Of course, they have lower resistivities than other soft ferrites but this drawback does not hamper their uses in the so called "low frequency" band (from about 10 kHz to 1 MHz).

The microstructure of this sort of ferrite is characterized by a mixture of spinel and inverse-spinel lattices. As a matter of fact, metal-ions are built into the crystalline lattice consisting of oxygen-ions. As a consequence, manganese-ions having high magnetic moments, increase the over-all value of system induction drastically. At the same time, zinc-ions having no magnetic moments, decrease anisotropy and thus contribute to the higher values of different permeabilities. As a result, macroscopical ferrite-properties are greatly improved (5).

To produce high quality manganese—zinc ferrites with excellent magnetic characteristics is a crucial task. On the one hand, a series of technological difficulties have to be overcome and on the other, conflicting requirements must often be met. A method to improve soft ferrites, in general, has been known since the early days of ferrite-research and the gist of it is to make them from raw materials as pure as possible. Assuming invariable processing technology, the quality of iron oxide, manganese carbonate and zinc oxide influences directly the initial permeability, losses, stability, etc. of the end-product. Consequently, the detection of impurities in raw materials, their quantitative determination as well as the techniques to eliminate them have become most important in development and research (6). According to this "traditional" way of furthering ferrite-quality even the water used during the technological process must be demineralized previously.

Appraising the value and importance of the right selection of ferrite raw materials, it must be recognized that this trend of development has brought about excellent results as seen by the very good magnetic properties of advanced manganese—zinc ferrites. On the other hand, possibilities in this field have been exploited almost totally — to such an extent that a "break-through", for instance, seems to be quite unlikely nowadays.

The second development trend for ferrites is closely connected with manufacturing technology. The relatively long and complex ferrite-making process includes a number of hidden traps for technologists who must take into account numerous process-parameters if they want to eliminate them. First of all, optimizing the sintering conditions is of utmost importance. Similarly, the temperature of preliminary and final sintering, the duration of heat treatment, the right value of partial oxygen-pressure as well as the use of an effective gas-atmosphere to protect the ferritizing process from harmful influences play crucial roles in ferrite-production. By masterful variations of these parameters, research-workers have succeeded, after innumerable experiments, to produce the highest quality ferrites of our days (4). In this respect, it is worth mentioning that e.g. initial permeabilities of 50 000 have been reached in single crystals, whereas loss-factors have been reduced to below 10^{-6}. Nevertheless, ferrite-

technology in itself cannot keep pace with the ever-growing requirements of electronic engineering. That is why the use of additives has come into the limelight of general interest. As a matter of fact, additives represent the third important development trend for manganese—zinc ferrites. Public attention has been focused on them as it has turned out that small amounts of various oxides (in most cases metal-oxides, of course) mixed in the prepared ferrite-charge have been able to improve considerably some of the magnetic properties of ferrites. A similar process has been witnessed in the field of permanent magnets where, an addition rare earth-metals to the traditional ferromagnetic alloys has brought about a real break-through in development, so much as that the limits of this rapid advance are not to be seen yet. (According to the added quantities, these rare earth-metals are used as real components and not additives, of course.) No doubt, additives as such have been known for a very long time in the field of soft ferrites, too, and since their introduction there have always been experiments with various oxide additions. Additives in ferrite-technology are called "magnetic enzymes" for their capacity of bringing about considerable changes in the ferrite microstructure in spite of their small amount. Correspondingly, structure changes alter some magnetic properties, sometimes improving and sometimes deteriorating them. It has been found, almost in all cases, that if a given additive has improved one or more properties, it quite similarly has been harmful to others. That is why there are very few additives recognized as generally useful.

Theory and experiments
to find useful additives for manganese—zinc ferrites

Under the conditions of the scientific-technological revolution of our days a rapid development of manganese—zinc ferrites is needed. First of all, their two most important magnetic characteristics, i.e. initial permeability and the loss-factor are to be improved, otherwise the ever-growing requirements (especially those of microelectronics) cannot be adequately met. As we have seen above, the traditional methods and techniques do not promise early results for major development, even the most promising trend that additives must be treated in a different way. According to a new conception, complex additives are found to consist more than one oxide. These oxides must be chosen in a way to counteract the harmful and enhance the good effects of one another.

Nowadays, extensive research-work is involved in the testing and checking of various (single and complex) additives as well as in studying their

effects on ferrite-structure to anticipate or even to define beforehand the desired magnetic properties. As regards manganese—zinc ferrites, additives are mainly needed to effect:

the decrease of the anisotropy-constant resulting in the increase of various permeabilities;

the increase of electrical resistivity leading to the reduction of losses.

Also, a number of other requirements are to be satisfied but as they are less important ones it seems to be quite superfluous to treat them in detail.

Instead, here is an example how additives should be tackled in general:

Let us assume that a given additive effectively lessens anisotropy in an Mn—Zn ferrite, to a high extent. It will be thus quite useless to mix this additive to the prepared ferrite-powder charge if, at the same time, the temperature-dependence of the anisotropy-constant increases. In reality, the additive must be completed by one or more other oxides able to counteract the deteriorating influence mentioned above. Otherwise, the final product will be lacking in temperature stability and earmarked as no good.

The changes in magnetic properties brought about by the various additives are consequences of structural changes in the ferrite material. An additive, however, can cause similar changes in two ways:

it can be built in the lattice-structure of the ferrite forming, in this case a one-phase homogeneous solid-state solution;

the additive itself may appear on the surfaces of ferrite-grains to create there a second phase.

By recent experiments with additives, it has been possible to determine the behaviour of a number of such oxides. For instance, CoO and TiO_2 belong to those penetrating into the lattice-structure, whereas CaO, SiO_2 and GeO_2 form a second phase on particle-surfaces. It seems quite peculiar that no experimental results about the behaviour of Ta_2O_5 have been published yet.

If high initial permeability of a manganese—zinc ferrite is required strictly, a one-phase structure is most likely to prove best. As a matter of fact, domain-wall shifting is effected relatively easily in such structures, especially if they contain few lattice-defects and pores. By the way, the number of lattice-defects can readily be reduced on surfaces by increasing the average grain-size.

Nevertheless, to produce low-loss ferrites, shortening the eddy-current paths is most effective and this in turn requires small average grain-size. Besides, a second phase is quite desirable, too, for forming an effective insulating layer at grain-boundaries. Of course, in a balanced state, the second phase should exhibit a much lower thickness than the domain-wall thickness.

It is quite clear from the above that the different claims can only be met by different structures often with contrasting qualities. That is why research-

workers are obliged to look for compromises and it has been found that such solutions are always to be based on grain-size control. Indeed, this latter is of utmost importance for modern ferrite-perfection.

Particle-size variation (i.e. growth and reduction) in ferrite-manufacturing depends in the first place on the sintering process. To investigate these phenomena more closely, let us consider ferrite samples after the complex ferritizing procedure during which the loose ferrite powder was transformed into solid parts. Grain-growth in these samples can be modelled as the movement of grain-boundaries in a system the free energy of which has gradually died down. Under this assumption, the velocity of the boundary movement corresponding to the resultant force is expressed by:

$$V = \sum_i v p_i ,$$

v being mobility (in the heat-activated process) and $\sum_i p_i$ is the resultant value of driving and impeding forces. As a rule, mobility depends on various diffusion processes that obey their own mechanisms. (E.g. diffusion may occur through the crystal-lattice, along grain boundaries, inside pores during the vapour-phase etc.) As for ferrites, the process complexity is further increased by the condition that anions and cations move at the same velocity to maintain balanced electrical charges. For testing purposes, ion-velocity is generally determined by means of the diffusion factor characterizing the slowest ions during diffusion along grain-boundaries. As a matter of fact, any additive whatever can accelerate particle-growth by increasing vacancy-concentration and at the same time diffusion velocity in the sub-lattice of the slowest ions. From this point of view, additive-valency and additive-solubility are of importance.

Some of the known driving forces are as follows:

Boundary energy. In general, low grain-size materials are recognized as having higher free energy amounts at grain-boundaries than the average (because of more lattice-defects on the surfaces).

Surface tension. Neighbouring crystallites differ from one another in orientation and thus in surface energy as well.

Discontinuous precipitation. The solid-state solution is over-saturated in front of the moving particle boundary, whereas a balanced two-phase system is prevailing behind it. The driving force is given by the difference between the two free energies.

Curvature of grain-boundaries. It is well-known that particle boundaries tend to move towards the particle centre, while the particles themselves may be

reduced so much that in the end they will disappear completely. Of course, at the same time the larger particles grow accordingly.

The impeding factors and forces acting against grain-growth are as follows:

a) The second phase and the pores. By reducing the free energy of grain-boundaries, they perform fixing effects.

b) Atoms of impurities dissolved into the ferrite-material. They hamper grain-growth according to various mechanisms.

c) Surface reactions of solid-liquid phases (3).

These phenomena are well-known from earlier observations and have been analyzed by a number of research-workers. Occasionally, it was possible to describe a few of them in detail by using the given parameters. Nevertheless, only some of the tendencies have been assessed and no general conclusions have been drawn yet. For the time being, detailed research and collecting experimental results should be carried out in this field.

Ferrite-research needs expensive and complex equipment and is very time-consuming, too. The results achieved in experiments by the use of various additives can be determined only after the length manufacturing procedure has been finished and there are already samples to be measured. We essentially strove to find a complex additive for our M2 F type manganese—zinc material as it was imperative to reduce losses, temperature-dependence as well as disaccomodation. According to our earlier investigations and information from literature, we intended to use an additive containing Ta, Ca and Si for the following reasons:

Ta_2O_5 had proved to be quite a promising oxide for enhancing loss-reduction first of all. Although tantalum is not at all cheap (1 kg Ta costs about 20 000 forints), we planned to use about 0.06 weight-percent of it. This amount, is still well below the prohibitive cost-limit.

As a matter of fact, CaO and SiO_2 are present as impurities in raw materials used in ferrite-manufacturing. On the average, $MnCO_3$ contains 0.05 weight-percent of CaO and the amount of SiO_2 in iron-oxide is 0.004—0.006 weight-percent. As is well-known in ferrite-technology, all additives have optimum ratios ensuring good effects on the magnetic properties of ferrites. Below or above these ratios the same additions usually prove to be harmful and deteriorate the magnetic characteristics. That is why we decided to determine the optimum amount of both CaO and SiO_2 and to clear up how they can be used to improve ferrite quality. Neither of them can be eliminated totally and therefore we always treated them together, i.e. as components of a two-member complex additive. We then wanted to continue our work by adding Ta_2O_5 to

the above two-component addition and to determine by a further series of experiments the new complex additive.

The above M2 F type manganese—zinc ferrite material is regarded as a high quality product used successfully in transmission engineering. At the beginning of our experiments its characteristics were as follows:

initial permeability: $\mu_i = 2200 \pm 20\%$
hysteresis losses: $h/\mu_i^2 \times 10^6 = 1.25$
disaccomodation: $D/\mu_i \times 10^6 = 6$
temperature-coefficient: $\alpha/\mu_i \times 10^6 = 0.5 \ldots 2.5$
loss-factor: $\tan \delta/\mu_i \times 10^6 = 8$.

After completion of our experiments, the improved M2 F type material was given the modified type mark of M2 FA.

Experiments and their results

To produce the necessary ferrite samples, only raw materials of high purity were used. The silicon and calcium content (see on page 23.) was never surpassed and alkali-impurities were strictly checked. In this respect, impurities having relatively large atomic diameters are especially harmful and greatly deteriorate the magnetic properties. According to theory, there is not enough room for them in the spinel-lattice which they thus distort, sometimes replacing it by a kind of a hexagonal structure containing even Na, K, Ca, Sr, Ba, etc. It is absolutely essential therefore that the amount of such impurities be reduced below an 0.01 weight-percent.

Basic materials of the following makes were used for our experiments:
iron oxide: Bayer WF 1352 Fe_2O_3 (German Federal Republic)
manganese carbonate: Usvico $MnCO_3$ (Japan)
zinc oxide: DAB—7 ZnO (United Kingdom).
The initial ferrite-composition was as follows:

	mole percent
Fe_2O_3	53.00
MnO	rest
ZnO	19.00
	100.00

The first milling, essentially a homogenizing operation, was accomplished in distilled water. This "wet" process lasted an hour during which the powder mixture was milled in an attritor-type ball-mill. Then the prefiring

(preliminary or first sintering) of the ferrite-powder followed in a laboratory furnace heated by supercantal elements. The first stage of prefiring at 900 °C lasted 2 hours, just like the second stage for which temperature was raised to 950 °C. Operating conditions of the second milling were the same as those during the first, although the milling time was now 2 hours. Then, after adding polyvinyl-alcohol to the mixture, it was granulated. Afterwards 30 toroid-cores of $\varnothing$ 28.5 × $\varnothing$ 16.3 × 20 mm were formed of the granular matter, by pressing. The final sintering of the samples was accomplished in a tunnel-furnace filled with a protective gas-atmosphere of $N_2 + 0.01\%$ O. The sintering temperature was 1290 ± 10 °C and this operation lasted 3 hours.

The produced core-samples formed 3 series as follows:

FA 1 cores:

They were made without any additives and served as reference cores having only the original M2 F type composition.

FA 2 cores:

These cores contained a complex additive composed of 0.030 weight-percent of CaO and 0.005 weight-percent of SiO_2 (the latter ingredient was originally present as an impurity).

FA 3 cores:

The complex additive they contained was composed of 0.075 weight-percent of CaO, 0.005 weight-percent of SiO_2 and 0.070 weight-percent of Ta_2O_5. (Again, SiO_2 was originally an impurity).

Experimental results

Five characteristic magnetic values of the samples were measured as follows:

initial permeability (μ_i)

hysteresis losses (h/μ_i^2)

the disaccomodation factor of permeability (D/μ_i)

the temperature coefficient of permeability (α/μ_i)

the specific loss-angle tangent $(\tan \delta/\mu_i)$.

Initial permeability was measured with a Maxwell-bridge and the same instrument was used to measure losses, too.

The following measuring frequencies were used:

20 kilocycles to measure initial permeability

100 kilocycles to measure $\tan \delta/\mu_i$

20 kilocycles to measure hysteresis losses.

Table 1

Measured magnetic characteristics of core-samples in series FA 1 (containing no additives)

Core-sample	μ_i	h/μ_i^2 $\times 10^{-2}$ m/A	D/μ_i	α/μ_i $\times 10^6/°C$	$\tan \delta/\mu_i$ $\times 10^6$
FA 10	2300	0.9	4.8	1.9	5.9
FA 11	2290	1.0	5.2	2.1	6.3
FA 12	2310	1.0	5.0	2.0	5.9
FA 13	2300	1.1	4.9	2.0	6.0
FA 14	2360	0.9	4.6	1.8	5.6
FA 15	2340	1.0	4.8	1.9	5.8
FA 16	2280	0.9	5.1	2.0	6.2
FA 17	2330	1.0	4.6	1.8	5.7
FA 18	2290	1.1	5.2	2.1	6.1
FA 19	2280	1.0	4.9	2.0	6.0

Useful impurities: 0.005 weight-percent SiO_2
in the iron-oxide
0.010 weight-percent CaO
in the manganese-carbonate.

To determine the temperature coefficient, the initial permeability was measured at 25 °C and at 60 °C. For measuring disaccomodation, the cores were charged with pulses of known decay having a duration of 40 seconds and a peak-value of 800 mA. Permeability variations in time were calculated by using the measured inductivity variations.

Experimental results are given in Tables 1—3.

Calculated average values and dispersions are shown Table 4.

Evaluation of results

According to the up-to-date concept of additives, all foreign matters present in the ferrite-material, i.e. impurities and additives, influence the magnetic properties of the end-product in a complex way. With this in mind, one can see from the test results at once that the complex additive containing Ta, Ca and Si effects the quality of the tested core-samples most favourably. The cores of series FA 3 are characterized by sufficiently high initial permeability, low losses and good stability factors representing the over-all aim of our development work. Although the samples belonging to series FA 2 and containing a complex additive of $CaO + SiO_2$ exhibit higher initial permeability than cores FA 3, they are set back by higher losses and lower temperature and time stability. Besides, the permeability dispersion of cores FA 3 is the lowest, quite favourable from the point of view of parameter-tolerances. The

Table 2

Measured magnetic characteristics of core-samples in series FA 2 (containing an additive composed of CaO and SiO_2)

Core-sample	μ_i	h/μ_i^2 $\times 10^{-2}$ m/A	D/μ_i	α/μ_i $\times 10^6/^\circ$C	$\tan \delta/\mu_i$ $\times 10^6$
FA 20	2470	1.3	4.4	0.5	5.3
FA 21	2540	1.3	4.5	0.8	5.7
FA 22	2530	1.4	4.7	0.7	5.8
FA 23	2500	1.2	4.4	0.6	5.6
FA 24	2480	1.2	4.3	0.8	5.4
FA 25	2460	1.3	4.4	0.8	5.2
FA 26	2500	1.4	4.5	0.8	5.6
FA 27	2510	1.3	4.7	0.7	5.6
FA 28	2500	1.1	4.6	0.7	5.7
FA 29	2480	1.2	4.4	0.6	5.6

Additive composition: 0.005 w% SiO_2
  0.030 w% CaO
  (impurities included).

Table 3

Measured magnetic characteristics of core-samples in series FA 3 (containing an additive composed of Ta_2O_5, CaO and SiO_2)

Core-sample	μ_i	h/μ_i^2 $\times 10^{-2}$ m/A	D/μ_i	α/μ_i $\times 10^6/^\circ$C	$\tan \delta/\mu_i$ $\times 10^6$
FA 30	2480	0.7	3.1	0.6	3.5
FA 31	2500	0.7	3.0	0.5	3.3
FA 32	2470	0.8	3.0	0.7	3.7
FA 33	2490	0.7	3.1	0.5	3.6
FA 34	2480	0.6	3.0	0.6	3.5
FA 35	2500	0.7	3.0	0.5	3.4
FA 36	2490	0.8	3.1	0.6	3.4
FA 37	2490	0.8	3.1	0.7	3.5
FA 38	2480	0.7	3.0	0.6	3.4
FA 39	2470	0.7	3.1	0.6	3.4

Additive composition: 0.005 w% SiO_2
  0.075 w% CaO
  0.070 w% Ta_2O_5*
  (impurities included)

* This amount has been determined in preliminary experiments.

Table 4

Calculated average values of magnetic characteristics from Tables 1—3 and their dispersions

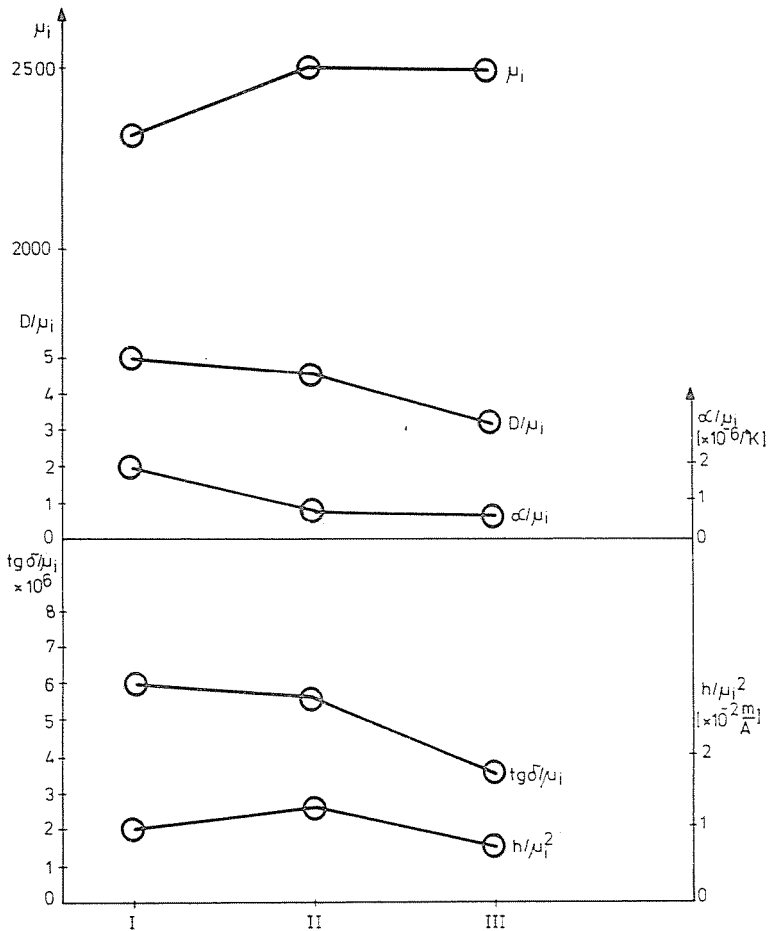| Series of core-samples	μ_i	h/μ_i^2 $\times 10^{-2}$ m/A	D/μ_i	α/μ_i $\times 10^6/°C$	$\tan \delta/\mu_i$ $\times 10^6$
FA 1	2300 ± 27	0.99 ± 0.07	4.91 ± 0.22	1.96 ± 0.11	5.95 ± 0.22
FA 2	2497 ± 25	1.27 ± 0.09	4.49 ± 0.14	0.70 ± 0.11	5.55 ± 0.19
FA 3	2495 ± 11	0.72 ± 0.06	3.05 ± 0.05	0.59 ± 0.07	3.47 ± 0.12

Fig. 1. Comparison of the measured magnetic characteristics of FA-type core-samples containing no additives and various complex additives, respectively. I: No additives, II. Additive containing SiO_2 and CaO, III. Additive containing SiO_2, CaO, Ta_2O_5

given experimental results agree well with those of Morisava—Okutani—Morita—Aojima (1). Investigating the effects of various tantalum additives, these authors succeeded to increase initial permeability, diminish core-losses and improve stability factors.

As for the complex $CaO + SiO_2$ additives, calcium silicate may theoretically be developed during heat-treatment. This compound, appearing as a second phase, forms a thin insulating coating on particle surfaces. As a consequence, initial permeability will increase. If, however, the rate of the two elements is not adequate for calcium silicate to be formed, the components of

Fig. 2. Electronmicrograph taken of the broken surface of an FA 3-type core-sample containing a complex additive of 0.005 w% SiO_2, 0.075 w% CaO, 0.070 w% Ta_2O_5

the complex additive may precipitate inside the grains causing lattice-distortions and deteriorating magnetic properties as well.

It is known from literature (2) that the upper limit of Ca + Si is about 0.3 w%. Above this, magnetic properties deteriorate rapidly. On the other hand, below this limit such additives enable to realize top-class ferrite-qualities equalling those made of pure raw materials. Some authors (6) contend that similarly small amounts of this complex additive promote sintering quite advantageously. Of course, the low increase in initial permeability experienced by us (Fig. 1) is still to be cleared up. For the time being, CaO and SiO_2 are regarded as useful impurities/additives up to a given limit.

Coming back to tantalum again, this metal is considered as very useful for ferrite perfection, all the same very little is known about the effect-mechanism by which its influence prevails. Nevertheless, it seems quite likely that tantalum gets built into the crystal-lattice of manganese—zinc ferrites and tantalum-ions substituting iron-ions in the lattice-sites alter and improve ferrite-quality (1).

In this connection electron-microscopic tests were accomplished to determine particle-sizes distributions in the samples. The electronmicrograph in Fig. 2 was taken from a sample-surface on the broken side of the core-sample. Accordingly, the average particle-size was about 30 μm. The quantity, distribution and forms of pores hint at near-optimum sintering conditions.

Conclusion

To further development, the optimum composition and amount of the used complex additive is to be determined. Again, loss-reduction and improving temperature and time stability are most desired (initial permeability seems to be adequate for the time being).

According to the concept of complex additives also other metal-oxides may be found to complete usefully the additive described above. A Japanese patent (1) refers to a ferrite-additive of five components to be used to produce manganese—zinc ferrites of "ultra low" losses. It claims a loss-factor of tan $\delta/\mu_i \times 10^6 = 1$. This practically means that the core-size can be further reduced for a new stage of miniaturization.

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