A FINE CARBONYL IRON POWDER FOR HIGH-FREQUENCY MAGNETIC POWDER MATERIALS

A. É. Fridenberg, V. G. Syrkin, I. S. Tolmasskii, and R. S. Fraiman

Moscow

Translated from Poroshkovaya Metallurgiya, No. 1 (13), pp. 33-41, January-February, 1963
Original article submitted December 21, 1961

For the production of magnetic powder materials meeting the requirements of present-day radio electronics and suitable for operation at frequencies above 50 Mc/sec, a carbonyl iron powder with particles 0.5-2 μ in size is required.

However, with the existing technology of preparation of this powder by the thermal decomposition of iron pentacarbonyl vapors at 300° in the presence of ammonia, it is only possible to produce two coarser fractions: one with particles 3-6 μ in size (class "R-4, grade 1", 80% of the whole powder) and another with particles 2-4 μ in size (classes "R-4, higher grade" and "R-2", 20% of the whole powder).

A very fine carbonyl iron powder, with particles 0.5-2 μ in size, can nevertheless be produced by the gas fractionation of the "R-4, higher grade" class powder in a separation installation consisting of a series of gradually decreasing cyclones.

The powder to be subjected to fractionation is charged into a feed hopper, from which it enters into a gas stream; during its travel from one cyclone to another, the required fractions are extracted from it by separation and collected in the containers of the three cyclones and a filter.

The fractions from the first two cyclones meet the requirements of the radio industry for the "R-4, higher grade" class powder provided that the cores made from them are prepared by the double insulation method.* The fractions from the third cyclone and the filter consist of particles 0.5-2 μ in size (80%) and have electromagnetic properties corresponding to the "R-2 extra" class: effective magnetic permeability μ_{ef} = 1.63-1.70, Q-factor ("goodness factor") Q_{rel} = 1.1, temperature coefficient of inductance (TCI) $\leq 50 \cdot 10^{-6}$ per 1°C.

This method has several disadvantages:

1. The preparation of the very fine powder is effected in an independent stage, which is separate from the main production process, i.e., decomposition of iron pentacarbonyl.

2. The feed hopper does not secure a sufficiently uniform supply of powder for separation, as a result of which the separation effect is reduced.

3. Nitrogen is the working gas and, consequently, a special system must be provided for its recirculation.

4. The process is not continuous, and the feed hopper must be periodically recharged.

The present investigation was undertaken with the object of developing a continuous method for the preparation of carbonyl iron powder, which would combine two processes: powder preparation by the decomposition of $Fe(CO)_5$ and separation of this powder in the actual decomposition system with the aid of a cyclone installation, in an atmosphere of the CO generated during the process. After a description and discussion of experimental data, the suitability of the resultant fine powder fractions for high-frequency magnetic materials will be examined.

* The double insulation method consists of the preliminary insulation of the powder with 0.2% of water glass [4% of a phenol-formaldehyde resin before compacting.

Continuous Method of Preparation of Fine Carbonyl Iron Powder

The processes of carbonyl iron preparation by the decomposition of $Fe(CO)_5$ and separation of the resultant powder were combined as shown diagrammatically in Fig. 1. As can be seen from the diagram, iron pentacarbonyl vapors are fed into the decomposition apparatus, after this has been blown through with nitrogen and carbon monoxide, where they dissociate in the presence of ammonia at 300-305° into iron and carbon monoxide. As a result of a five-fold increase in the volume of the system, a strong directional stream of carbon monoxide is produced; this stream entrains the fine iron particles which have not settled down in the container of the decomposition apparatus and carries them out to the group of cyclones.

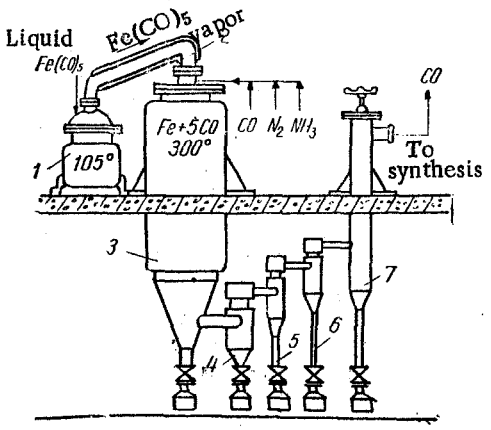

Fig. 1. Diagram of apparatus for production of fine carbonyl iron powder: 1) vaporizer (height 1 m, diam 0.5 m); 2) preheater (length 2 m, diam 0.2 m); 3) decomposer (height 3 m, diam 0.75 m); 4) first cyclone (height 0.5 m, diam 0.15 m); 5) second cyclone (height 0.4 m, diam 0.12 m); 6) third cyclone (height 0.3 m, diam 0.10 m); sleeve-type beta-filter (height 2.5 m, diam 0.25 m).

The powder fractions settling in the containers of the three cyclones represent specific, gradually decreasing particle sizes. The finest fractions are discharged from the third cyclone and the beta-filter. After passing through the filter, the powder-free outlet gases are pumped to the iron pentacarbonyl synthesis plant for further utilization.

As can be seen from this description, the cyclone group has been included between the decomposition apparatus and the beta-filer of the standard, existing carbonyl-process installation. In the experiments, the consumption of liquid iron pentacarbonyl was 10.5-11.5 liters/hr, while the ammonia feed was maintained at a level of 40 liters per 1 liter of $Fe(CO)_5$. The sizes of the individual units are given in Fig. 1. The duration of the individual operations exceeded one day. The powder was discharged every 3 h during the process.

Tables 1 and 2 give the experimental data obtained in this investigation, including the test conditions, materials balance sheets of the carbonyl iron powders produced, and sieve analyses of the powders.

In order to provide a comparison of powders produced by the existing and new methods, Table 3 lists the physicochemical and electromagnetic characteristics of the new powder.

As can be seen from Table 1, the yield of the fine fraction (from the third cyclone and the beta-filter) amounted to 5-6% of the total quantity of powder obtained from the cyclone group and the filter (i.e., the total quantity of the "R-2" and "R-4, higher grade" powders produced by the existing procedure). In the first operation, this yield was 8.1%. These figures are in full agreement with the results obtained in [1], in which the yield of the required fraction was 5-8% of the charge. The weight of the discharge from the first cyclone ranged between 14.2 and 25.5%, which was due to the increased feed of iron pentacarbonyl.

The weights discharged from the second and third cyclones were approximately constant in all operations. With increasing feed, the yield of powder from the second cyclone rose slightly, the values obtained in the three experiments being 3.8, 6, and 7.2%, respectively; the yield of powder from the third cyclone was more or less constant, and amounted to 1.3-1.6%. It follows from this that percentage yield from the first and second cyclones depends on iron pentacarbonyl feed: the greater the feed, the greater is the amount of powder carried out from the decomposition apparatus and settled in these cyclones.

As regards the third cyclone and the filter, it has already been pointed out that the yield of powder from their containers remains constant (1.3-1.6%) irrespective of changes in the rate of feed of iron pentacarbonyl (10.5, 11.0, and 11.5 liters/hr). This feature is particularly important, because it shows that production of fine fractions with a given sieve analysis by this method is stable irrespective of the rate of iron pentacarbonyl feed, and consequently, that the method is flexible.

For a detailed study of the separation process, the carbonyl iron powders discharged from the individual cyclones and the filter were subjected to particle size analysis. The results of these analyses are given in Table 2. The pro-

TABLE 1. Powder Preparation with Simultaneous Extraction of Fine Fractions

Test No.	Discharge No.	$Fe(CO)_5$ feed	Pressure in decomposer, mm Hg	Weight of discharged powder, g						% of yield relative to total powder produced					Weight of powder in all cyclones and filter, g	Powder from third cyclone and filter as % of this sum
				decomposer	first cyclone	second cyclone	third cyclone	filter	total	decomposer	first cyclone	second cyclone	third cyclone	filter		
1	1	10,5	10	7700	1020	160										
	2		10	8250	1830	530										
	3		10	7870	1650	440										
	4		10	678	900	280	600	0								
	Total			30600	5400	1400	600	0	38010	81	14,2	3,8	1,6	0,0	7410	8,1
2	1	11,0	20	8140	1650	400										
	2		40	8440	1650	360		330								
	3		40	7320	2180	750										
	4		40	10100	2300	700										
	5		45	8000	1840	570										
	6		40	9920	1840	850										
	7		40	8750	1750	450										
	8		50	9030	1960	1000	1320	530								
	Total			60700	16170	5800	1320	860	84130	72	19,8	6,0	1,6	1,0	23430	6,0
3	1	11,5	50—20	10970	2430	950										
	2		20—40	10800	2700	1100										
	3		40—15	11600	3650	1120		40								
	4		15—40	10700	3000	1180		50								
	5		35—45	12490	500	1510		20								
	6		45—25	10430	9000	1460		15								
	7		25—15	4170	2150	500	1400	250								
	Total			71160	27930	7820	1400	375	108785	66	25,5	7,2	1,3	0,35	37525	5,0

cedure employed involved the statistical calculation of the number of particles visible under the microscope at a magnification of 1350 diameters.

It will be seen from Table 2 that the specific powder fraction with particles 1-2 μ in size is the easiest to separate by the new technique, its sieve analysis being more uniform than that of the remaining fractions obtained by separation.

TABLE 2. Sieve Analysis of Resultant Powders

Point of discharge	Test No.	Discharge No.	Amount of particles of given size as % of total quantity					Percentage of conglomerates in total quantity	Mean diam., μ
			≤1 μ	2 μ	3 μ	4 μ	≥5 μ		
First cyclone	1	1	51,96	22,97	12,42	7,26	5,28	41,55	2,70
		2	54,86	23,76	9,52	6,67	5,09	30,84	2,72
		3	48,52	23,24	15,38	6,52	8,20	38,02	2,76
		4	40,65	28,64	17,45	6,65	6,53	33,96	2,92
Second cyclone	1	1	57,70	28,50	9,40	2,70	1,70	26,10	2,12
		2	52,71	23,70	8,85	3,61	1,14	28,64	2,09
		3	50,47	32,58	10,42	5,29	1,16	27,13	2,25
		4	65,32	22,47	7,26	3,13	1,25	29,46	2,02
	3	5	41,36	21,36	17,65	6,85	2,38	28,41	2,60
Third cyclone	1	4	89 57	8,44	1,69	0,30	0,00	8,83	1,24
	3	1—7	71,77	21,81	4.5	1,48	0,50	.22,39	1,77
Filter	3	1—7	90,67	6,38	1,85	0,60	0,05	4,56	1,13

Thus, in the powder collected in the third cyclone and the filter, the total quantity of particles 1-2 μ in size is 97%. The analysis of the powder from the third cyclone and the filter differs substantially, particularly by its small content of particles 4-5 μ in size (under 1%), from that of the powders discharged from the first and second cyclones.

Table 3 shows the electromagnetic characteristics of separated carbonyl iron powder. As can be seen from the table, at a frequency of 50 Mc/sec, the powders discharged from the third cyclone have an effective magnetic permeability of μ_{ef} = 1.74-1.84 and a Q-factor of Q_{rel} = 1.1, which corresponds to the "R-2 extra" class being introduced at the present time. The powder which is currently being produced for operation at a frequency of 50 Mc/sec has Q_{rel} = 0.9 and μ_{ef} = 1.45 ("R-2" class). Thus, the new method gives an increase of Q_{rel} from 0.9 to 1.1 and of μ_{ef} from 1.45 to 1.84, the average increase in both cases being 12%. The marked decrease of Q_{rel} in the powder from the beta-filter is due to the extreme fineness of this powder (about 1 μ).

At a frequency of 5 Mc/sec, the value of μ_{ef} gradually decreased from the first to the third cyclone, being 3.11-3.16 (first cyclone), 2.97-3.07 (second cyclone), and 2.84-2.91 (third cyclone). The goodness factors of the powders were 2.07-2.22 for the first cyclone, 2.05-2.16 for the second cyclone, and 2.10-2.18 for the third cyclone, i.e., increased with decreasing particle size.

As can be seen from Table 3, the chemical composition of the powder in all cyclones was relatively constant. It may thus be stated that the changes in the electromagnetic properties of the powders between the cyclones were due solely to changes in their particle size, resulting from gas fractionation in the cyclone group located immediately after the decomposer.

In order to examine the feasibility of prolonged operation, test No. 3, with an over-all duration of 25.5 h (not counting unscheduled discharging), was carried out. During the process (Table 1), the pressure in the decomposer occasionally rose by 10-40 mm Hg, but subsequently returned to normal. When the plant was dismantled and examined, it was found that this was due to the periodic blocking of a tube with carbonyl iron powder, which was cleared when the pressure in the system rose to 40-50 mm Hg.

It may thus be considered that prolonged operation of the proposed system is feasible.

Electromagnetic Properties of "R-2 Extra" Class Carbonyl Iron

In order to evaluate the advantages of the new, "R-2 extra" class powder against the "R-2"class powder at present in production, a study was made of the electromagnetic properties of the former powder. The physicochemical properties of these powders are quoted in Table 4.

TABLE 3. Physicochemical and Electromagnetic Characteristics of Powders

Point of discharge	Test No.	Discharge No.	Mean diam. μ	Chem. composition, %		$f = 5$ Mc/sec		$f = 50$ Mc/sec	
				C	N	Q_{rel}	μ_{ef}	Q_{rel}	μ_{ef}
First cyclone	1	1	2,7	0,66	0,70	2,11	3,12	0,77	1,91
		2	2,72	0,66	0,69	2,04	3,11	0,77	1,89
		3	2,76	0,65	0,69	2,08	3,10	0,79	1,91
		4	2,92	0,69	0,71	2,07	3,16	0,72	1,93
	2	2	2,81	0,74	0,72	2,08	3,10	0,70	1,85
		5	2,72	0,72	0,73	2,01	3,04	0,63	1,84
		7	2,82	0,68	0,73	2,01	3,12	0,72	1,83
Second cyclone	1	1	2,12	0,70	0,71	2,14	2,99	0,96	1,85
		2	2,09	0,65	0,71	2,13	3,00	0,94	1,86
		3	2,25	0,69	0,70	2,16	2,97	0,95	1,85
		4	2,02	0,67	0,72	2,16	2,98	0,97	1,85
	2	1—4	2,03	0,71	0,71	2,05	3,07	0,96	1,79
		4—8	2,20	0,68	0,71	2,05	3,12	0,97	1,85
	3	2	2,10	0,76	0,71	2,08	3,01	0,99	1,77
		5	2,60	0,67	0,68	2,16	2,96	0,96	1,79
		7	2,03	0,74	0,73	2,08	3,03	0,99	1,76
Third cyclone	1	1—4	1,24	0,69	0,70	2,16	2,91	1,10	1,84
	2	3—8	1,37	0,71	0,69	2,18	2,84	1,01	1,74
	3	1—7	1,48	0,64	0,64	2,18	2,85	1,16	1,73
Filter	3	3—6	1,13	0,76	0,70	2,09	3,13	0,84	1,86
		7	1,09	0,76	0,71	2,06	3,11	0,88	1,86

TABLE 4. Physicochemical Properties of "R-2" and "R-2 extra" Class Powders

Powder type	Particle size, μ	Impurity content, %		
		C	N_2	O_2
"R-2"	2.3	0.78	0,77	0.81
"R-2 extra"	1.19	0.81	0,78	0.87

TABLE 5. Electromagnetic Characteristics of Powders at Frequency of 50 Mc/sec

Powder type	Mean particle diameter, μ	Q_{rel}	μ_{ef}
"R-2 extra"	1.3	1.12	1.70
"R-2"	2.3	0.88	1.73

Cylindrical and toroidal cores were made from these powders. Double insulation, as described above, was used in their manufacture. For applying the first insulating layer, the iron powder was placed in a mixer into which a solution of water glass in distilled water was poured during continuous stirring. The mixture was stirred for 30 min at room temperature and subsequently heated during continuous stirring, to 100°. Stirring went on until the mixture was completely dry, after which it was cooled to 20°.

For depositing the second insulating layer, an alcohol solution of bakelite resin was poured into the mixer during continuous stirring. The amount of ethyl alcohol was 12-15% of the powder weight. The mixture was stirred for 30 min at room temperature, after which it was heated to 30° during continuous stirring until it was completely dry. After cooling to room temperature, the insulated powder was compacted into cores of the required shape. The cores were held in air for 4 h and then heated for 1 h at 130°.

For evaluating the electromagnetic characteristics of the new magnetic powder material against those of the old type at a frequency of 50 Mc/sec, cylindrical cores of the STsG-2 type were made and tested according to VTU standard 1024-54 [2]. In order to determine the electromagnetic characteristics of the new magnetic powder material in the frequency range up to 250 Mc/sec on annular cores (external diameter 35 mm, internal diameter 25 mm), measurements were made, using a Czechoslovak high-frequency resonance bridge, of initial permeability μ_{i_1} and the tangent of the magnetic loss angle, tan δ_μ, as functions of frequency at different field intensities. The losses were broken up into constituents by the method described in [3]. For obtaining frequency dependence curves, tan δ_μ and μ_{i_1} were measured with the aid of high-frequency parameters at several predetermined frequencies.

TABLE 6. Electromagnetic Characteristics of Powders at Frequencies up to 250 kc/sec

Powder type	μ_{i_1}	$\delta h_r \cdot 10^3$	$\delta f_{i\tau} \cdot 10^9$	$\delta h_i \cdot 10^3$	$TC\mu_{i_1} \cdot 10^6$
"R-2"	11.4	0.22	2.35	0.16	64
"R-2 extra"	9.15	0.05	0.79	0.07	31

Note: μ_{i_1} – initial permeability; δh_r – coefficient of Rayleigh hysteresis losses; $\delta f_{i\tau}$ – overall coefficient of eddy current and viscosity losses; δh_i – coefficient of initial hysteresis losses; $TC\mu_i$ – temperature coefficient of initial permeability.

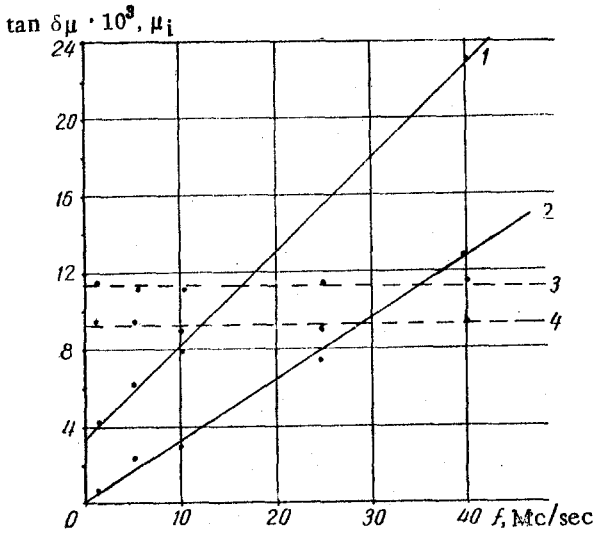

Fig. 2. Effect of frequency on tangent of magnetic loss angle and initial permeability of "R-2" (curves 1 and 3) and "R-2 extra" (curves 2 and 4) powders: curves 1 and 2 – tan δ_μ; curves 3 and 4 – μ_i.

Table 5 gives values of the goodness factor Q_{rel} and the effective permeability μ_{ef} of specimens of the two magnetic powder materials, measured at a frequency of 50 Mc/sec.

It can be seen from this table that the relative goodness of the magnetic material from the "R-2 extra" powder is approximately 30% higher than that of the material from the "R-2" powder, at practically identical permeability.

Table 6 shows the electromagnetic characteristics of the two powders, measured in the range up to 250 kc/sec. It can be seen from Table 6 that, compared with those of "R-2" powder, the loss components of "R-2 extra" powder are much smaller as a result of the use of very fine iron particles.

Figure 2 shows the effect of frequency on the tangent of the magnetic loss angle and initial permeability of the two powders. As can be seen, the electromagnetic properties of the new powder in the frequency range investigated are higher than those of the "R-2" class powder.

SUMMARY

1. A method has been proposed for the continuous preparation of very fine fractions of carbonyl iron powder, which combines the processes of iron pentacarbonyl decomposition and separation of the resultant powders by the outlet gases.

2. Powder production with the simultaneous extraction of very fine fractions was investigated.

3. It is shown that the fraction obtained from the third cyclone, which consists of particles 0.5-2 μ in size, has electromagnetic characteristics which satisfy the "R-2 extra" class requirements.

4. The yield of "R-2 extra" powder amounts to 5-6% of the total amount of the "R-4, higher grade" powder which is entrained by outlet gases from the decomposer apparatus.

5. It was established that, compared with the "R-2" powder currently produced in the Soviet Union, "R-2 extra" carbonyl iron powder has improved electromagnetic characteristics in a wide frequency range.

LITERATURE CITED

1. R. S. Fraiman, V. G. Syrkin, and V. L. Volkov, Khim. Prom. 7 (1961).
2. VTU standard No. 1024-54, Ministry of Chemical Industry, USSR. Carbonyl Iron Powder for Radio Engineering [in Russian] (1954).
3. K. M. Polivanov, Ya. N. Kolli, and L. P. Soboleva, "Permeability and losses of magnetic powder materials," Izvest. Akad. Nauk SSSR, 23, 3 (1959).
4. L. I. Rabkin, High-Frequency Ferromagnetics [in Russian] (Moscow, State Physicomathematical Literature Press, 1960).
