United States Patent

[72] Inventor   Alfred M. Laing
                 Butler, Pa.
[21] Appl. No. 714,805
[22] Filed   Mar. 21, 1968
[45] Patented   Sept. 21, 1971
[73] Assignee   Spang Industries, Inc.

[54] PROCESS OF MAGNETIC PARTICLE
     PREPARATION
     11 Claims, No Drawings

[52] U.S. Cl.. **148/104,**
                                    148/31.55, 148/105
[51] Int. Cl....................................... **H01f 1/24**
[50] Field of Search.. 148/100,
          104, 105, 31.55; 75/170; 252/62.55; 117/100 M

[56]   References Cited
          UNITED STATES PATENTS

1,943,115	1/1934	Ellis............................	148/104
2,110,974	3/1938	Gillis...........................	148/31.55 X
2,531,445	11/1950	Laycock......................	148/31.55 X
2,937,964	5/1960	Adams et al.................	148/105
2,977,263	3/1961	Harendza-Harinxma	148/104
3,014,825	12/1961	Harendza-Harinxma	148/104
3,255,052	6/1966	Opitz...........................	148/105

Primary Examiner—L. Dewayne Rutledge
Assistant Examiner—G. K. White
Attorney—Shanley and O'Neil

ABSTRACT: Magnetic particle cores suitable for phase shifting applications having a permeability substantially above 275 and core losses below 0.24 ohms per henry per unit of inductance at 1,800 Hertz are produced by a novel combination of steps including one or more of the following: reduction of prepowder anneal additives, selective distribution of magnetic particle sizes, adding flexibility to the electrical insulation applied to the magnetic particles, elevated pressure compacting, surface etching of the pressure compacted core after hydrogen anneal, and an oxidation heat treatment following surface etching. Surface etching and oxidation combine to increase the breakstrength and improve other properties of various permeability cores, for example, from 125 through 300 permeability.

PROCESS OF MAGNETIC PARTICLE PREPARATION

This invention relates generally to preparation of magnetic particles for compacting into magnetic components and, more particularly, to high permeability particle cores exhibiting low core losses in the audio frequency and related frequency 5 ranges, and to methods of manufacturing such cores.

A basic process for manufacture of particle cores is disclosed in the U.S. Pat. No. 2,105,070 to Bandur. The process steps include the preparation of magnetic particles, electrically insulating the particles, compacting into a magnetic core, 10 and annealing the core. This basic process has been capable of producing molybdenum-containing permalloy particle cores having a permeability of about 125 and having core losses acceptable for audiofrequency applications.

Attempts to improve on this process have been numerous 15 and continuous since its origin. The problem is to increase the permeability without destroying usefulness of the core due to an increase in core losses. Successful improvements of core properties have generally resulted from treatment of the compacted core, for example by boiling, solution treatment, and 20 reanneal of the compacted core as disclosed in the U.S. Pat. No. 2,977,263 to Harendza-Harinxma issued Mar. 28, 1961, No. 014,825 issued Dec. 26, 1961, and No. 3,132,952 issued May 12, 1964.

The primary objective of particle core improvement en- 25 deavor is to increase the permeability of such cores while maintaining core losses within acceptable limits. Particle cores are used in electrical circuits operating at voice frequencies and related frequencies up to about 20,000 cycles per second where low core losses are a major consideration. Also these 30 cores find certain applications at much higher frequencies which further accentuate the importance of low core losses. In the audio frequency and related frequency ranges, components approaching theoretical perfect reactance are desirable in order to obtain high quality performance. In filters, for 35 example, shorter cutoff, better defined resonance, and higher attenuation ratios are realized with high quality inductors. Quality in inductors can be assessed from the "Q" factor. The "Q" of inductors is defined as a ratio of reactance to re- 40 sistance:

$$Q = \frac{2\pi f L}{R_{DC} + R_{AC}}$$

where
f = frequency in cycles per second
L = inductance in henries, and
R_{DC} = wire resistance in ohms
R_{AC} = resistance (ohms) due to losses of the core, including eddy current losses, hysteresis losses, and residual losses.

Provided core losses can be kept within industry specifica- 50 tions, there are a number of reasons why increasing the permeability is important, for example, savings on core material, savings on wire, and improved circuit design because of smaller components. As is well known in the art, efforts to date to improve on metallic particle cores have resulted in 55 commercially available cores with acceptable core losses having an upper limit of 200, and slightly higher, permeability. Special laboratory techniques may yield acceptable cores of about 240 permeability; however, it has not been possible to produce a particle core having a permeability above 275 with 60 core losses acceptable in the industry for the uses considered above.
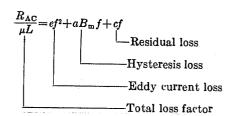
The present invention teaches novel procedures for preparation of magnetic powders for magnetic components and, specifically, preparation of molybdenum-containing per- 65 malloy powders for producing magnetic cores of higher permeability, lower core losses, increased mechanical break-strength, more linear temperature characteristic, and better moisture resistance. These teachings enable commercial production of molybdenum-containing permalloy powder 70 cores having a permeability substantially above 275 and having core losses within industry specifications.

Core losses are reflected in the windings of a core as resistance losses and constitute loss of energy in an inductor. Core losses, as used in the core manufacturing art, include 75

eddy current losses which vary directly with the square of the frequency, hysteresis losses which vary directly with the flux density and the frequency, and residual losses which vary directly with the frequency.

These core losses may be expressed by the V. E. Legg formula covered in Legg's paper entitled, "Magnetic Measurements at Low Flux Densities Using the Alternating Current Bridge" from the Bell System Technical Journal of Jan. 1936. This formula is as follows:

$$\frac{R_{AC}}{\mu L} = ef^2 + aB_m f + cf$$

Residual loss
Hysteresis loss
Eddy current loss
Total loss factor

R_{AC} = effective (total measured) resistance minus DC resistance
μ = permeability
L = inductance (henries)
f = frequency (Hz)
B_m = flux density (gausses)
e = eddy current loss coefficient
a = hysteresis loss coefficient
c = residual loss coefficient To meet industry standards, cores must have a total core loss no higher than 0.240 units (ohms per unit permeability per unit inductance) at 1,800 Hertz (cycles per second), with a core loss of 0.200 units at 1,800 Hertz being the accepted average.

To meet these standards and improve magnetic and mechanical properties as well, several novel steps are combined in the present invention. These steps will be described in relation to production of novel high permeability molybdenum-containing permalloy powder cores with acceptable core losses. Previously, the highest permeability core, of the type discussed, available commercially was a "200 perm" core. The present teachings make the "300 perm" core commercially available. Of special significance is the fact that these "300 perm" cores have losses acceptable within industry standards for audiofrequency uses, i.e. below 0.240 units.

Molybdenum-containing permalloy is an alloy consisting essentially of nickel, iron and molybdenum. In practice, the standard alloy contains about 2 percent molybdenum, about 81 percent nickel and the balance iron. However, alloy constituents can vary within the following percentages without serious detriment to properties:

1.68 –4.0%	molybdenum,
78.0–83.0%	nickel, and
the balance iron	

Methods for pulverizing the alloy are known, e.g. the U.S. Pat. No. 1,669,649 to Beath et al. In practice the metallic constituents of the alloy are melted together and additives which embrittle the alloy are made in the molten state. This treatment facilitates a fine crystalline structure in the solidified alloy that enables reduction to a fine powder by conventional rolling, grinding, and pulverizing techniques.

In departing from prior practice, the invention teaches selection of a variety of particles sizes to obtain an optimum packing factor, i.e. an optimum density of magnetic powder and optimum space for electrical insulation (electrically equivalent to air space) in the compacted product.

A typical particle size distribution to produce 300 perm cores in accordance with the process of the present invention is:

TABLE I

Average Particle Size Microns	Sieve Mesh-size*	% by Weight
90	from −120 to +230	10–15%
65	from −230 to +400	25–35%
37 (or less)	−400	45–65%

* −120 to +230 covers particle sizes which will pass through a 120 mesh screen but will not pass through a 230 mesh screen using normal sieve practice.

The important concept discovered here is that the optimum packing factor to obtain high permeability and acceptable core losses is not obtained by use of single particle size screening but rather by selective screening and distribution of particle sizes. The high permeability—acceptable core loss product of the invention can be obtained by selecting particles with about one part by weight having an average particle size of 90 microns, about three parts by weight having an average particle size of 65 microns, and about six parts by weight in which the average particle size is no greater than about 37 microns. These proportions can be broadened to emphasize certain core properties and changes in the electrical insulation also permit variation in these proportions. However, a larger overall-average particle size tends to increase both permeability and core losses while smaller overall-average particle size tends to decrease both permeability and core losses.

After pulverizing and sieving, the powder is annealed to relieve the strains induced during brittle practice, that is during the production of the powder. To prevent welding of the particles during this anneal, additions of nonagglomerating material must be blended with the powder. Such material must remain nonreactive or inert at powder annealing temperatures. In the prior art, preanneal additions constituted about 0.3 percent to 1.0 percent by weight of the metal powder. An important discovery of the present invention relates to better use of the limited amount of distributed nonmagnetic gap available in producing the higher permeability product of the present invention; that is, this space can be better used to provide more effective electrical insulation of the particles rather than being occupied by preanneal additives—when proper steps are followed. By the procedures of the present invention, the preanneal additive is drastically reduced to about 0.02 percent to about 0.05 percent by weight of the metallic powder; preferably such additives are held below about 0.03 percent by weight. Typically ceramic clays such as talc or kaolin are added to prevent agglomeration; a preferred preanneal additive is powdered kaolin.

The subsequent powder anneal is held to a temperature of about 1,250° F. for about 1 ½ hours in a nonoxidizing atmosphere, e.g. an atmosphere containing free hydrogen. Temperatures significantly higher than about 1,250° F. are avoided in order to eliminate agglomeration of the metal particles. With the present invention it is possible to avoid the 1,400° F. to 1,600° F. powder anneals of the prior art without sacrificing electrical properties. In fact, a higher permeability—low core loss product is obtained.

During the powder anneal the water of crystallization of the kaolin, which constitutes about 13 percent by weight of this preanneal additive, is driven off. The kaolin should be in the uncalcined condition before the powder anneal since it has been found that calcined kaolin is not as effective as standard kaolin in preventing agglomeration of the metal particles.

After annealing, the work product is sieved through a 50 to 100 mesh screen to remove any lumps which may be formed;

however, this does not change the analysis of the metal powder sizes which have been selected to produce the desired packing factor. This sieving merely breaks up loose lumping which may occur.

The magnetic powder is then electrically insulated utilizing a slurry including, by present practice, a silicate, an inert metallic oxide, and a ceramic clay. For example, a solution containing about 67 grams of sodium silicate, about 100 grams of milk of magnesia, and about 6,000 cc. of deionized water is prepared to insulate about 50 pounds of powder. The electrical insulation is applied in a plurality of coats with the first coat ordinarily not including a ceramic clay additive in order to utilize the preanneal nonagglomerating additive present with the metal powder. Subsequent coats after the first coating, utilize about 1,400 cc.'s of the above solution with about 22 grams of powdered kaolin added. In preferred practice the electrical insulation is applied in four separate coats with intermediate dryings of the coatings being carried out at temperatures up to about 315° F. The total electrical insulation, dry weight, is less than about 0.4 percent by weight of the metal powder weight.

Highly beneficial results are obtained by the addition of a plasticizer coating during the insulation process. Plasticizer as used herein, refers to a material for imparting flexibility to the electrical insulation or a major ingredient of the electrical insulation, e.g. the metallic silicate in the disclosed insulation. As taught by the present invention, the plasticizer must maintain this capability of imparting flexibility to the electrical insulation during processing steps up to and including the compacting step.

Preferably the plasticizer is added as a final coating to the insulated particles; however plasticizers exist which require intermediate coating for best results. Suitable plasticizers of the latter category include starches, sugars, or glycerin and a wetting agent. A plasticizer suitable for adding as a final coating is ammonium lignosulfonate in a liquid carrier.

The purpose of the plasticizer is to impart flexibility to the electrical insulation and permit higher than usual pressures during compacting of the particles while avoiding mechanical cracking of the electrical insulation. In accordance with the teachings of the invention, the plasticizer should maintain its capability of imparting flexibility during the temperatures encountered in applying electrical insulation and those encountered in compacting. Preferably the plasticizer should be driven off during the high temperature core anneal or, at least, not impair the insulation or leave a reaction product having reduced electrical insulation properties.

After the insulation process, including the use of a plasticizer, the insulated powder is sieved through a 50 to 100 mesh screen to remove lumps and chips of insulation. This sieving is carried out without changing the basic magnetic particle sieve analysis.

The insulated powder is pressed into cores at a pressure which is significantly higher than that previously specified for molybdenum-containing permalloy powder cores. The compacting pressure taught by the present invention for the production of higher permeability cores is ordinarily in the range of about 135 to 150 tons per square inch and preferably is about 140 tons per square inch. The plasticizer makes the insulation more flexible and reduces compacting friction.

Without a plasticizer, the core losses increase considerably at the higher compacting pressures taught. A portion of the decrease in core losses available with the present invention can be traced to the decrease in surface welding stemming from the decrease in compacting friction. The reduction in core losses also stems from decreasing mechanical breakage of the electrical insulation between particles which apparently existed in the prior art practice. Further, the plasticizer reduces the amount of lubricant needed in pressing.

However there are limits to the amount of plasticizer which can be safely used since mechanical breakstrength of the core

decreases rapidly above certain low level percentages. When ammonium lignosulfonate is used with an insulation containing sodium silicate, the amount of dry plasticizer should be about 0.06 percent by weight of the metal powder weight.

The invention includes discovery of a step to maintain desired mechanical breakstrength of the finished product. The coaction of this step, which will be described later, offsets any weakening effect on the cores caused by the plasticizer so that cores with mechanical breakstrength equivalent to prior art cores, without plasticizer, can now be made notwithstanding the use of a plasticizer.

After pressing, the cores are annealed between about 1,000° F. and about 1,500° F., preferably about 1,250° F. for approximately 40 minutes in a nonoxidizing atmosphere, for example, an atmosphere containing pure hydrogen. The cores are quenched in water after removal from the annealing furnace.

Practice of the process of the invention described thus far consistently produces cores within the normal tolerance range for 300 permeability cores, however during production runs core losses can vary beyond accepted standards. To commercially produce cores with losses consistently within industry specifications, that is below 0.240 units, the invention teaches use of a surface etching step. By surface etching is meant removal of the skin effect resulting from present-day compacting techniques used in commercial production of molybdenum-permalloy cores.

The cores are surface etched subsequent to the hydrogen anneal which follows pressing. The cores should not be surface etched prior to this anneal. In general, chemical etching is preferred in order to avoid adding any mechanical strains to the particles. A typical etching practice utilizes a 50 percent nitric acid solution with an etching time of 20 seconds, plus or minus 5 seconds with temperature maintained at 80° F. ± 5° F. An alternate etching procedure is approximately 3 minutes in 40° Baume nitric acid with temperature maintained at 80° F. ± 5° F.

Surface etching can cause a slight decrease in permeability but this decrease is limited to about 0.5 percent to about 5 percent of the core permeability. Typically, a 302.1 permeability core may be reduced to 300.6 permeability and a 323.3 permeability core may be reduced to 317.3. However, core losses decrease at a much greater rate than permeability; decreases in core losses up to about 50 percent are typical. For example, the above 302.1 permeability core had a $\Delta R/\mu L$ value of 0.117 before surface etching. This core loss was reduced to 0.0972 by surface etching. The above 323.3 permeability core had a $\Delta R/\mu L$ value of 0.413 units before etching which was reduced to 0.203 units, more than 50 percent, by surface etching. In brief, while the permeability may be decreased as much as 5 percent by surface etching, the core losses are reduced as much as 50 percent.

While 300 permeability cores with core losses within accepted standards can be produced consistently in production utilizing the above steps, the invention also includes discovery of a novel step in the treatment of compacted cores which further improves electrical and mechanical properties of molybdenum-containing permalloy particle cores. This step coacts with other steps in the production of 275 and higher permeability cores. For example, this step helps increase the mechanical strength of a high permeability core offsetting any weakening effect of a plasticizer coating. This step also acts to offset the effects of the higher pressures used in producing 275 and higher permeability cores by decreasing core losses which could result from such higher pressures. However, this novel step also improves the mechanical and electrical properties of lower permeability cores as well.

In accordance with the invention, after annealing of the powder cores in a nonoxidizing atmosphere such as hydrogen, and after the surface etching, the cores are heat treated in an oxygen-containing atmosphere, for example, air. The order of these two heat treatments, that is the hydrogen anneal and the air heat treatment, cannot be reversed without loss of the

benefits obtained by annealing in hydrogen, followed by surface etching, followed by oxidation. It is believed that a hydrogen anneal subsequent to the heat treatment in air reduces the bonds formed during oxidation.

The oxidation step should be carried out at a temperature between about 600° F. and about 1,000° F. for an interval of about 10 to 15 minutes. A preferred oxidation treatment is applied at about 850° F. for about 15 minutes. It should be understood that this oxidation treatment has a time-temperature relationship, that is, a longer period of time, for example, 1 ½ hours at a lower temperature, for example about 225° F., can be utilized to provide similar oxidation, but the time involved is uneconomical and can have slightly detrimental side effects on other properties. In general, within the above limits, the improvement in breakstrength is greater at higher temperatures.

Oxidation increases the breakstrength of the core as much as 75 percent depending on the particular core, decreases total core losses as much as 25 percent (chiefly a decrease in eddy current losses), and markedly decreases the effects of moisture on a core.

Certain benefits of oxidation heat treatment are more pronounced with high permeability cores. A 300 permeability toroidal core, unoxidized, having a 1.06-inch outer diameter, about 0.5-inch inner diameter, and 0.44-inch height, breaks at a 170 pounds of force per square centimeter of radial cross sectional area. An otherwise identical core, oxidized at about 850° F. for 12 minutes, breaks at 260 pounds per square centimeter, an increase in breakstrength of 50 percent. The breakstrength, however, of a 115 to 135 permeability core of similar size shows an increase in breakstrength of about 10 percent when treated in the same fashion.

Breakstrength measurements are made in accordance with the industry accepted "Vertical Core Breakstrength Test." This is a mechanical test in which force is applied on diametrically opposite sides of a painted core's outer diameter with maximum tangential contact being made on both sides. The ramming force required to break the core is measured in pounds per square centimeter of a radial cross section of the core.

This test provides an important parameter for designating a mechanical characteristic, that is the breakstrength, of the product of the present invention. If the breakstrength measured in accordance with the "Vertical Core Breakstrength Test" is plotted versus cross-sectional areas of a radial segment of cores in square centimeters a linear relationship is found to exist. The minimum acceptable value of this ratio of pounds of breakstrength to area of a radial segment in square centimeters, for a painted core, is 290. For example, a core with an outer diameter of 1.06 , an inner diameter of 0.580 inch, and a height of 0.440 inch has a radial section area of 0.635 square centimeters. The minimum accepted breakstrength of a painted core of this size is 184.15 pounds. The breakstrength factor of a core having this minimum breakstrength would be 184.15 divided by 0.635 which equals 290. The electrically insulating paint applied to the exterior as a final step in processing particle cores may be conventional, e.g. an enamel core paint with a thickness of roughly 7 to 12 mils.

All molybdenum-containing permalloy cores from 125 permeability to 300 permeability show a substantial decrease in core losses after the oxidation heat treatment taught. The higher permeability cores show best results when annealed in the range of roughly 750 to 850° F. for about 12 minutes. However with all cores of the type described, if the temperature of the oxidation heat treatment is allowed to rise above about 1,000° F., the cores will deteriorate with regard to losses.

Oxidation helps solve a problem of long standing in this art, that is the detrimental effect of humidity on permeability; see "Stability Characteristics of Molybdenum Permalloy Powder Cores" by C. D. Owens, "Electrical Engineering," Mar. 1956, pages 252–255. Past efforts have been concentrated on finding and applying coatings and packings for cores which would

7

stop the detrimental effect of humidity. These efforts, from the point of view of practical handling problems and economics, have been at the limits of their capability for some time. The oxidation step taught by the invention helps to solve this problem in the core itself and, for the first time in this art, brings the humidity problem under practical and economic control.

Humidity decreases the permeability of a core. The oxidation treatment taught by the invention reduces this decrease in permeability by at least 50 percent in all cores and by greater amounts in the higher (300) permeability cores disclosed. For example, an unoxidized 300 permeability core, with conventional electrical insulating enamel coatings of about 7 to 12 mils total thickness on its exterior surface, shows a change of −3.3 percent in permeability when exposed to 95 percent relative humidity in air at 150° F. for 5 days. Otherwise identical cores, oxidized between 575° F. and 850° F. for 12 minutes had a permeability change of −1.0 percent. Under the same conditions 200 permeability cores unoxidized showed a −2.2 percent change in inductance in this test while the oxidized cores showed an average change of −1.1 percent in permeability.

Also the effect of changes in temperature on permeability, i.e. the "change in permeability versus change in temperature" characteristic, is made more linear. This is partially due, it is believed, to a reduction in the effect of the difference in coefficients of expansion between the paint on a core and the core itself, especially at lower temperatures. Evidently an oxidized core is better able to withstand the force of contraction of the paint on the core because of the increased breakstrength resulting from oxidation.

Improvement in breakstrength due to oxidation is especially beneficial with the higher permeability cores compacted from electrically insulated particles having a plasticizer coating. A plasticizer dry coating weight of 0.06 percent to 0.1 percent ammonium lignosulfonate significantly improves core losses but decreases the breakstrength of such cores slightly. Surface etching and heat treating in air cause equivalent or higher breakstrength than that experienced with conventional cores of the same size without a plasticizer.

The following table lists permeability and core losses, obtained in commercial production of 300 permeability core (normal tolerance ±8 percent permeability), for the various standard core sizes, using the teachings of the present invention.

TABLE II

OD (in.)	ID (in.)	Ht (in.)	μ	$\Delta R/\mu L$
0.250	0.110	0.110	281	0.170
0.310	0.156	0.125	307	0.121
0.400	0.200	0.156	308	0.120
0.500	0.300	0.187	310	0.190
0.900	0.550	0.300	277	0.130
1.060	0.580	0.440	298	0.128
1.300	0.785	0.420	292	0.130
1.410	0.880	0.412	298	0.146
1.570	0.950	0.570	308	0.210

In describing specific embodiments of the invention, detailed steps, values, and determinations have been set forth

8

which not only enable practice of the invention but also provide guidelines form modification of the specific embodiments by those skilled in the art, therefore the scope of the invention is to be determined from the following claims.

1. Process for preparing magnetic material for compacting into a magnetic component comprising the steps of
   providing finely divided magnetic powder
   blending the magnetic powder with a nonagglomerating material which is substantially nonreactive at annealing temperatures for the magnetic powder, the nonagglomerating material comprising about 0.02 to about 0.05 percent by weight of the magnetic powder.
   annealing the magnetic powder blended with nonagglomerating material in a nonoxidizing atmosphere,
   electrically insulating the annealed magnetic powder with the total dry weight of electrical insulation comprising less than 0.40 percent by weight of the magnetic powder, and
   adding a plasticizer to impart flexibility to the electrical insulation, the plasticizer being nondecomposing at the temperatures required for applying the electrical insulation.

2. The process of claim 1 in which the magnetic powder consists essentially of molybdenum, nickel, and iron in alloy form.

3. The process of claim 1 in which the magnetic powder is blended with about 0.02 percent to about 0.03 percent by weight uncalcined kaolin before annealing the magnetic powder and the annealing is carried out at a temperature of about 1,250° F.

4. The process of claim 1 in which an electrical insulation coating is applied to the annealed powder from a slurry including a metallic silicate and an aqueous suspension of an inert metallic oxide.

5. The process of claim 1 in which the magnetic powder is electrically insulated by applying a plurality of coatings of a slurry including a metallic silicate, an aqueous suspension of an inert metallic oxide, and a ceramic clay with intermediate drying of each coating and in which the plasticizer coating is applied to a dried electrical insulation coating.

6. The process of claim 1 in which the electrical insulation is applied in a plurality of individual coatings and in which the plasticizer is added with an electrical insulation coating.

7. The process of claim 2 further including the step of compacting the electrically insulated magnetic powder with plasticizer at a pressure of about 135 tons to about 150 tons per square inch to form a magnetic core.

8. The process of claim 7 further including the step of annealing the compacted magnetic core between about 1,000° F. and about 1,500° F. in a nonoxidizing atmosphere.

9. The process of claim 8 further including the step of surface etching of the annealed magnetic core to minimize skin welding effect resulting from pressure compacting.

10. The process of claim 9 further including the step of oxidizing the surface etched magnetic core by heating the magnetic core above about 225° F. but not higher than about 1,000° F. in an atmosphere containing free oxygen.

11. The process of claim 10 in which the surface etched magnetic core is heated in the atmosphere containing free oxygen to a temperature above about 600° F.

65

70

75

UNITED STATES PATENT OFFICE
CERTIFICATE OF CORRECTION

Patent No. 3,607,462   Dated September 21, 1971

Inventor(s) Alfred M. Laing

It is certified that error appears in the above-identified patent and that said Letters Patent are hereby corrected as shown below:

Col. 1, line 24, "No. 014,825" should read --

No. 3,014,825 --

Col. 2, line 56, "1.68-4.0%" should read --

1.6—4.0% --

Signed and sealed this 11th day of April 1972.

(SEAL)
Attest:

EDWARD M.FLETCHER,JR.   ROBERT GOTTSCHALK
Attesting Officer   Commissioner of Patents