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INSULATING COMPOSITIONS FOR LAMINATIONS AND PRODUCT PRODUCED THEREWITH

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This invention relates to compositions suitable for application to ferrous metal surfaces to produce insulating coatings thereon, the process of applying the compositions and the product produced therewith.

In preparing laminations of magnetic material for the building of magnetic cores therefrom, it is necessary that the laminations be provided with electrical insulation between one another in order to minimize eddy currents and to achieve low losses. The insulation should preferably be an extremely thin film integral with the surfaces of each lamination to provide for a high space factor. In many cases, it is further necessary that the insulating material applied to laminations should withstand the elevated temperatures which are encountered in strain annealing cores after assembly in order to improve their efficiency. Temperatures encountered in stress relieving occasionally reach 1200° C. and are rarely less than 600° C. In addition, it is highly desirable that the insulation for the magnetic sheet material be capable of withstanding bending, scraping and other mechanical abuse or chemical treatment which may be encountered in forming the magnetic material to shape and assembling cores therefrom. Magnetic material is usually cut or punched after the insulating material is applied thereto, and thereafter the cut or punched magnetic material may be bent or wound in order to produce a predetermined core structure. Additionally, the assembled cores may be subjected to machining or grinding, and, in some cases, etching with acids in order to remove burrs and the like.

It has been proposed heretofore to treat the surfaces of ferrous magnetic laminations with phosphoric acid in order to produce an insulating iron phosphate film or coating on the surfaces of the laminations. Extensive experience with phosphoric acid treated ferrous magnetic material of this kind has shown that the insulation resistance of the films is relatively low, often of the order of 1 to 2 ohms per square centimeter when tested by simple contact pressure alone. When the test contacts are applied with a force of 50 pounds per square inch upon the phosphoric acid treated material and twisted, the insulation resistance frequently drops to values of the order of 0.15 to 0.5 ohm per square centimeter. Often the phosphate film is disrupted during twisting and the resistance is zero. For numerous applications, the resistance values of less than 1 ohm per square centimeter are too low to be satisfactory. Even in small trans-

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formers an interlaminar resistance of at least one ohm per square centimeter is necessary to limit eddy current loss in the core to a workable value. The lack of adherence and abrasion resistance as evidenced by decrease of resistance upon twisting of the contacts as well as visual observation of the breaking up of the film indicates that the insulation produced by phosphoric acid alone reacting on iron is not satisfactory for many uses. Considerable difficulty has been encountered in actual experience with this type of insulation on laminations.

Tests have shown that the addition of a powdered refractory such as magnesia, alumina, or silica flour to the phosphoric acid before application to ferrous metal does not materially improve the insulating film produced. The ohmic resistance of the films is not significantly greater than that produced with the phosphoric acid alone. The adherence of the phosphate films is not better when embodying such refractory solids. Therefore, phosphoric acid carrying powdered refractory solids does not produce films having high mechanical strength, adherence and high resistivity.

The object of this invention is to provide a tenaciously adherent insulating film possessing high ohmic resistance on ferrous metal surfaces.

A further object of the invention is to provide a composition composed of phosphoric acid and a silica hydrogel capable of producing adhered films of high ohmic resistance on ferrous material by heat treatment.

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

According to the present invention, aqueous compositions are prepared containing a relatively high phosphoric acid concentration and a substantial amount of a silica hydrogel substantially all of whose particles are from 1 to 20 microns in diameter. Upon applying a coating of the compositions to the surface of ferrous metal and heat treating at temperatures of 300° C. to 950° C., films of high adherence and of extraordinary ohmic resistance are produced. This phenomenon is particularly unexpected since compositions of silica flour of a fineness of 300 mesh added to phosphoric acid will not produce films that have properties comparable with the properties of films produced with the present composition.

More specifically, the compositions of the present invention are composed of 100 parts by weight of orthophosphoric acid, from 5 to 55 parts by weight of a silica hydrogel composed of particles

substantially all of which are in the range of from 1 to 20 microns in diameter and from 7 to 15 parts by weight of water for each part of the silica hydrogel, the maximum amount of water not exceeding 600 parts by weight. The composition may be conveniently prepared by admixing 100 parts by weight of aqueous orthophosphoric acid solution having more than 50% orthophosphoric acid by weight and from 60 to 200 parts by weight of an aqueous silica hydrogel dispersion, prepared as will be described hereinafter, containing from 10 to 15% by weight of silica in the form of particles substantially all of which are from 1 to 20 microns in diameter.

A silica hydrogel suitable for the practice of the present invention is made by mixing sodium silicate, preferably having a ratio of from 3 to 3½ SiO₂ groups per Na₂O group, with sulphuric acid. The proportions of sulphuric acid and the sodium silicate are approximately equimolecular, an excess of acid being preferred. The mixture, which is known as a hydrosol, is permitted to age, for example, in tubs holding several hundred pounds of the hydrosol. From 3 to 5 hours aging results in a gelation of the hydrosol to a colloidal hydrogel of the desired consistency, and the hydrogel is emptied from the tubs and broken into small lumps as by passing over a screen. The broken hydrogel is thoroughly washed with water to remove any excess acid, sodium sulphate and any other water-soluble salts resulting from the reaction. After being thoroughly washed, the hydrogel particles containing approximately 80% moisture are passed through a colloid or ball mill with additional water in about a similar amount to that present being introduced. A thixotropic, creamy dispersion is produced which may have a maximum of approximately 15% silica in the form of hydrogel particles though, for practical uses, approximately 11% silica is the optimum that forms a stable, workable dispersion. The dispersion can be readily diluted with water. The hydrogel portion of the dispersion consists of particles substantially all of which are from 1 to 20 microns in diameter. Apart from the water, the hydrogel particles are substantially pure silica, analysis (water-free) showing better than 99% silica. A small amount, approximately 0.001%, of ammonia may be added as a stabilizing ingredient.

For treating ferrous metal the coating composition prepared from this dispersion should contain more than 10% by weight of phosphoric acid. The water introduced with the phosphoric acid and the silica hydrogel dispersion may be supplemented with additional water, though best results have been obtained with a minimum of water being present.

The composition may be applied to ferrous metal and ferrous base alloys of various kinds: pure iron, silicon-iron alloys having from 1% to 7% silicon, or higher, iron-nickel alloys having up to 50% nickel, iron-cobalt alloys and low carbon iron. The metal may be of strip, sheet rod, wire or other form, or in the shape of stamped punchings.

The composition of this invention is preferably applied as a thin coating in such proportions as to provide from ½ to 5 pounds of the silica in the hydrogel proper for each 300 square feet of metal surface. Particularly good results have been obtained with from 1 to 2 pounds of silica for each 300 square feet of metal surface.

After application of the composition to the metal surfaces, the metal and applied coating

are heat-treated at a temperature of from 300° C. to 950° C. for a length of time sufficient to drive off the water and to cause a reaction of the composition ingredients with the metal surface to take place. The precise nature of the reaction is not known but smooth homogeneous coating less than one-quarter of a mil in thickness results. The time of the reaction need only be a few seconds at temperature of 700° C. and higher. At lower temperatures, the reaction may be somewhat longer. Several minutes heat treatment at temperatures from 650° C. to 950° C. has given good results in practice. It is preferred to maintain a relatively low oxidizing or inert atmosphere in the furnace to prevent undue oxidation and the formation of rust. In a strong oxidizing atmosphere such as air, heating should not be conducted for more than two minutes at 600° C. With a relatively non-oxidizing or inert atmosphere, for example nitrogen gas, or nitrogen-hydrogen mixtures such as are usually used in annealing, the coatings may be heat-treated for considerable lengths of time. In case the ferrous metal strip is to be reannealed subsequently, it has been found desirable that the heat-treating temperature of the strip with the applied composition should approximate the reannealing temperature.

The following examples set forth compositions which have been applied to silicon iron laminations 13 mils thick.

Example I

A composition was prepared by admixing 10 gallons of 75% phosphoric acid and 100 pounds of silica hydrogel suspension containing 11 pounds by weight of silica proper. This mixture formed a stable suspension of coating consistency. Rolls of 13 mil thick sheet silicon iron 14 inches wide were roller-coated with the suspension, a total of 9600 pounds of silicon iron being coated, and passed into an oven having an atmosphere at a temperature of 760° C. The sheets were in the oven for approximately 1½ minutes. The sheets were found to be uniformly coated with a hard film of insulating refractory. The sheets were tested with a C-clamp tester whose construction and operation are described in detail in the Horstman et al. Patent 2,354,123, issued July 18, 1944. The tester at no time indicated a resistance of below 100 ohms per square centimeter, and indicated infinity resistance for nearly all the spots tests on sheets produced. Such resistance was recorded even when the tester was twisted several times.

Example II

A composition was prepared by admixing:

	Parts
Water	100
Phosphoric acid 85%	70
Silica hydrogel dispersion (11% SiO ₂)	70

The composition was applied to silicon iron laminations in the proportion of one gallon to 320 square feet of surface. Laminations with this composition were baked at 700° C. for 15 seconds. The coatings so produced were extremely adherent and indicated infinity resistance.

Example III

A composition was prepared by mixing:

	Parts
Water	100
Phosphoric acid 85%	100
Silica hydrogel dispersion (11% SiO ₂)	100

When applied to laminations in the same proportions as in Example II and baked at 700° C. for 15 seconds, a tenaciously adherent coating of infinite resistance was produced on ferrous metal sheets.

Example IV

A composition was prepared by admixing:

	Parts
Water	100
Phosphoric acid 85%	100
Silica hydrogel dispersion (11% SiO ₂)	150

The composition when applied to silicon iron laminations and heat treated at 700° C. for 30 seconds produced films having infinite ohmic resistance. The films were smooth and very adherent.

Example V

A composition was prepared by admixing the following:

	Parts
Water	100
Phosphoric acid 85%	100
Silica hydrogel dispersion (11% SiO ₂)	250

This composition applied to silicon iron sheets and baked for 30 seconds at 650° C. yielded a hard, tightly adherent coating of infinite ohmic resistance as determined by the C-clamp tester above identified.

The compositions of this invention have numerous advantages. The aqueous compositions are highly stable and will not precipitate, or deteriorate even on long-continued storage, as for several weeks. They do not contain flammable solvents, such as are present with ethyl silicate compositions, and, therefore, are safe for use with any type of furnace. The compositions may be diluted indefinitely with water without breaking; therefore, the thickness of the applied coatings may be controlled readily by the amount of water added.

Silicon iron laminations coated with the insulating film derived on heat treating the compositions of this invention on their surfaces are refractory at elevated temperatures. Tests of magnetic cores produced therewith which were annealed at 700° C. showed that practically no laminations stuck together after the annealing operation.

Since certain changes in carrying out the above process and certain modifications in the composition and members produced therefrom may be made without departing from the scope thereof, it is intended that all matter contained in the above description shall be interpreted as illustrative and not in a limited sense.

We claim as our invention:

1. A composition suitable for applying to ferrous metal to produce on the metal after heat treatment tenaciously adherent insulating coatings having a high ohmic resistivity, composed of, in combination, 100 parts by weight of orthophosphoric acid, from 5 to 55 parts by weight of a silica hydrogel having from 10% to 15% by weight of silica, the hydrogel composed of particles substantially all of which are in the range of from 1 to 20 microns in diameter, and from 7 to not more than 15 parts by weight of water for each part of the silica hydrogel, the maximum amount of water not exceeding 600 parts by weight, all combined as an aqueous dispersion.

2. A coating composition in the form of an aqueous dispersion composed of 100 parts by weight of aqueous orthophosphoric acid having more than 50% phosphoric acid and from 60 to 200 parts by weight of an aqueous silica hydrogel dispersion containing from 10% to 15% by weight of silica, the hydrogel particles substantially all of which are from 1 to 20 microns in diameter.

3. In the process of treating ferrous metal to provide on the surface thereof a tenaciously adherent film of high ohmic resistance, the steps comprising applying to the surface of the ferrous metal a composition composed of 100 parts by weight of orthophosphoric acid, from 5 to 55 parts by weight of a silica hydrogel composed of particles substantially all of which are in the range of from 1 to 20 microns in diameter, and from 7 to not more than 15 parts by weight of water for each part of the silica hydrogel, the maximum amount of water not exceeding 600 parts by weight, the composition being applied to provide from ½ to 5 pounds of the silica in the hydrogel for each 300 square feet of metal surface, and heat-treating the ferrous metal and applied composition at a temperature of from 300° C. to 950° C. to drive off the water and cause a reaction of the composition with the ferrous metal to take place to provide the adherent film.

4. Magnetic sheet material comprising a body composed of ferrous magnetic metal and a surface film composed of an adherent refractory coating, the coating being derived by applying to the surface of the ferrous metal a composition composed of 100 parts by weight of orthophosphoric acid, from 5 to 55 parts by weight of a silica hydrogel composed of particles substantially all of which are in the range of from 1 to 20 microns in diameter, and from 7 to not more than 15 parts by weight of water for each part of the silica hydrogel, the maximum amount of water not exceeding 600 parts by weight, the composition being applied to provide from ½ to 5 pounds of the silica in the hydrogel for each 300 square feet of metal surface, and heat-treating the ferrous metal and applied composition at a temperature of from 300° C. to 950° C. to drive off the water and cause a reaction of the composition with the ferrous metal to take place to provide the adherent film.

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