Patented Apr. 21, 1953

2,635,995

# UNITED STATES PATENT OFFICE

2,635,995

## ELECTROPHORETIC INSULATING COATING

Henry T. Swanson, Lancaster, Pa., assignor to Radio Corporation of America, a corporation of Delaware

No Drawing. Application June 19, 1948, Serial No. 34,156

4 Claims. (Cl. 204—181)

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The invention relates to insulating coating materials and more particularly to such materials containing aluminum nitrate and the process of making the same.

The cataphoretic process of applying an insulating coating to a metal base has heretofore been used when the configuration of the base has made the use of the drag or spray process impractical. For example, coiled heater filaments for indirectly heated cathodes are preferably coated by the cataphoretic process since the drag or spray process might leave inner surfaces of the filament inadequately covered.

Some coating materials employed in the past for application to a base by cataphoresis have included aluminum oxide, commercial aluminum nitrate, magnesium nitrate, water, and an alcohol such as methanol. The aluminum oxide is usually the preponderant constituent of the solids portion of the coating, the quantities of aluminum nitrate and magnesium nitrate being variable and their relative amounts being determined by the desired thickness of the coating. The nitrates provide the required negative radical of the electrolyte, which includes water and an alcohol such as methanol. The aluminum oxide, aluminum nitrate, and magnesium nitrate acquire electrical charges when the solution is subjected to a potential difference at displaced portions thereof by suitable electrodes, one of which may constitute the metal base to be coated. After the metal base is coated it is baked at a relatively high temperature, for example, about 1600° C. to drive off the liquid constituent thereof and to convert the magnesium nitrate and aluminum nitrate to the oxides which possess good insulating properties.

One of the difficulties associated with prior procedures involving a cataphoretic application of a coating of the kind described has been that the finished coating shows the presence of pits and a number of radial cracks therein, which result in a peeling or chipping of the coating from the base. There appeared to be no known way of preventing or controlling this undesirable condition of the coating with the result that it has been difficult, if not impossible, to obtain a coating having predictable characteristics.

Another difficulty resides in the lack of stability of the coating material, which heretofore has made it necessary that the coating material be used in a relatively short time after its preparation.

An object of the invention is to provide an insulating coating material of improved insulating and adherence characteristics.

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It is another object to provide an insulating coating having a relatively smooth surface, free from cracks.

A further object is to provide an improved cataphoretic process for applying an insulating coating to a base.

Another object is to provide a novel form of aluminum nitrate which will be a contributing factor in obtaining a relatively stable coating solution and a consequent uniformly smooth and adherent coating material.

An additional object is to provide a coating formulation with reproducible end results.

Still another object is to provide a coating which can be depended upon for close control of thickness of insulating material.

Further objects and features will become clear as the description continues.

According to the invention an insulating coating material is provided which may include the constituents before referred to, with the exception that the commercial aluminum nitrate is treated in a special manner so that its inclusion in the coating will render the coating free from chipping or peeling and make the coating material more stable for a substantially longer period of time than heretofore possible.

The treatment of the aluminum nitrate referred to comprises subjecting commercial aluminum nitrate which has the chemical designation of $Al(NO_3)_3.9H_2O$ to a plurality of heat treatments in distilled water wherein the heat is kept below the temperature at which the aluminum nitrate will yield aluminum oxide. The yield of this treatment is aluminum nitrate and aluminum hydroxide which has a non-crystalline structure. The heat treatments referred to are repeated until the yield has an aluminum oxide equivalent of from 20% to 33%. These heat treatments constitute a process by which the formula for commercial aluminum nitrate is changed to a value such that the resultant material is chemically equivalent to from 20 to 33% by weight of aluminum oxide.

This relationship between aluminum oxide and aluminum nitrate can best be shown by the following theoretical consideration.

$Al(NO_3)_3.9H_2O$ is equivalent to, or for present purposes is potentially capable of producing, aluminum oxide ($Al_2O_3$) to an extent of about 13.6% by weight. Aluminum nitrate—$Al(NO_3)_3$ without any water of crystallization is chemically equivalent to about 24% $Al_2O_3$ by weight. Aluminum hydroxide ($Al(OH)_3$) is chemically equivalent to 65.4% aluminum oxide ($Al_2O_3$) by weight. Thus the formulation most suitable for provid-

ing between 20 and 33% aluminum oxide equivalent may be anhydrous aluminum nitrate with some further decomposition of part of this material to aluminum hydroxide or some similar compound plus nitric acid which later is eliminated by the processing.

The decomposition would take place in accordance with the following reaction:

$$Al(NO_3)_3 + 3H_2O \longrightarrow Al(OH)_3 + 3HNO_3\uparrow \quad (1)$$

or

$$2Al(NO_3) + Heat \longrightarrow Al_2O_3 + 3N_2O_5\uparrow \quad (2)$$

Heating commercial aluminum nitrate

$$(Al(NO_3)_3.9H_2O)$$

as outlined in the above processes gives rise to the following reactions, which take place simultaneously:

$$Al(NO_3)_3.9H_2O + Heat \longrightarrow Al(NO_3)_3 + H_2O\uparrow \quad (3)$$

$$Al(NO_3) + 3H_2O + Heat \longrightarrow Al(OH)_3 + 3HNO_3\uparrow \quad (4)$$

Thus the processing primarily involves a reduction of water and nitric acid content of the aluminum nitrate and the formation of some aluminum hydroxide which tend to stabilize the solution and supply the proper conditions for proper deposition of the insulating material.

It can be seen from the equations shown above that it is possible to eliminate only the water of crystallization from commercial aluminum nitrate $Al(NO_3)_3 \cdot 9H_2O$ and reach a formula corresponding to an aluminum oxide content within the range specified. Thus a single heat treatment as described above would be sufficient to produce the formulation prescribed. This is but a step in the right direction, however, since single evaporation has not been sufficient in practice for best results. The subsequent heat treatments serve to produce aluminum hydroxide and nitric acid, the latter being eliminated according to the reaction indicated in Equation 4. The extent of the reaction required for practising the invention may be determined by actual plating tests. Although my experience has been that more than one evaporation should be employed, it is possible that by proper control of the rate of evaporation one treatment would suffice.

It can be seen readily that a number of theoretical combinations of $Al(NO_3)_3$, $H_2O$, and $Al(OH)_3$ could be obtained with an equivalent of from 13.6% to 65.4% $Al_2O_3$. One thing is certain, however, as can be readily seen from the above relationships, in order to produce a material which is equivalent to more than 24% $Al_2O_3$ it is necessary to have some decomposition of $Al(NO_3)_3$ to $Al(OH)_3$ and $HNO_3$ (nitric acid) which latter is driven off by the heat treatment. According to the invention, the primary requisite is that the $Al_2O_3$ equivalent be greater than 20%. Best results are obtained with materials potentially capable of producing or chemically equivalent to from 20% to 33% $Al_2O_3$. It is possible, however, and theoretically probable, that material equivalent to more than 33% aluminum oxide ($Al_2O_3$) could be used satisfactorily.

Aluminum oxide ($Al_2O_3$) is used as a means of comparison mainly because the simplest analytical procedure for control of these materials involves converting them to this product in accordance with the following reactions:

$$2Al(NO_3)_3.9H_2O + Heat \longrightarrow Al_2O_3 + 3N_2O_5\uparrow + 9H_2O\uparrow$$

$$2Al(NO_3)_3 + Heat \longrightarrow Al_2O_3 + 3N_2O_5\uparrow \quad (6)$$

$$2Al(OH)_3 + Heat \longrightarrow Al_2O_3 + 3H_2O\uparrow \quad (7)$$



These equations represent standard methods of analysis. The heat required in these cases is that of the Bunsen burner. Arrows pointing upward indicate that the compound so marked is driven off by the heat.

By way of example, the heat treatment of the invention may be carried out by placing about 0.5 lb. commercial aluminum nitrate

$$(Al(NO_3)_3.9H_2O)$$

in a large evaporating dish and heating for about thirty-five minutes at a temperature below 135° C. but allowing the material to approach this temperature as the evaporation proceeds. The material initially becomes liquid at the end of fifteen minutes but reverts to its solid non-crystalline state after an additional twenty minutes have elapsed. In this state the material may be said to have a syrupy consistency. During the heating step constant stirring of the mixture is necessary and the frequency of the stirring should be increased when the material begins to solidify.

To the solid material is then added from 300 to 400 cc. of distilled water, in which the material is caused to dissolve by a gentle heating thereof. The temperature is then raised to a value below 135° C. until the water has evaporated and the material has returned to the solid and somewhat syrupy state. This step is repeated once more, after which the material is subjected to a test to determine its aluminum oxide equivalent. If the aluminum oxide equivalent is less than 20% of the treated aluminum nitrate, the step referred to must be repeated until this value of aluminum oxide equivalent, or a value not exceeding 33%, is secured. It is then submitted to a practical test designed to function as a control of suitability for use.

The test employed to determine the suitability of the processed aluminum nitrate for the sake of convenience is one of actual application in a coating suspension. It consists of preparation of a small quantity of coating solution and application of the insulating material in a regular fashion. If the result is a smooth coating free from cracks before and after firing, the prepared nitrate is approved for use. If the coating is rough or cracked further treatment of the nitrate is necessary.

The invention produces a new material adapted to serve as a constituent of an insulating coating and having the desirable properties of imparting to the coating material a stability of conductivity resulting from a desired value of concentration of hydrogen ions at a desirable level per unit of volume. The reduction in the amount fo water of crystallization of aluminum nitrate from 9 molecules per molecule of aluminum nitrate to a lower value accomplished by the invention, eliminates a source of water having a tendency to hydrolyze to nitric acid during storage of the material. Nitric acid reduces the stability of the coating material and its elimination, or at least substantial reduction, results in a relatively stable solution.

A further feature of the invention concerns the relative amounts of magnesium nitrate and the treated aluminum nitrate heretofore described, required in an insulating coating for best results. For example, a coating material according to the invention may include 2358.7 grams Alundum, 1620 cc. of a solvent such as denatured alcohol, 1500 cc. of distilled water, 47.25 grams of magnesium nitrate, and an

amount of aluminum nitrate determined by the following formula:

$$\text{Weight of aluminum nitrate} = \frac{13.95}{C}$$

where C equals the aluminum oxide equivalent expressed as a fraction of the aluminum nitrate which should lie between the extremes of .20 to .33. The numerator 13.95 is a constant and represents the aluminum oxide equivalent of aluminum nitrate in grams to be used with 47.25 grams of magnesium nitrate.

Various modifications may be made in the invention without departing from its spirit and it is intended to include them within the scope of the appended claims.

I claim:

1. Method of preparing aluminum nitrate for use in a relatively stable solution for application as an insulating coating comprising heating commercial aluminum nitrate to drive off at least part of the water of crystallization content thereof and to convert a portion of said aluminum nitrate to aluminum hydroxide, and until the treated aluminum nitrate and the derived aluminum hydroxide contain an aluminum oxide equivalent of from 20 to 33% by weight.

2. An electrophoretic insulating coating material comprising aluminum oxide, a solvent, magnesium nitrate and aluminum nitrate having an aluminum oxide equivalent of from 20 to 33% by weight, the amount of said aluminum nitrate being related to the amount of said magnesium nitrate and the aluminum oxide equivalent of said aluminum nitrate, the amount of said aluminum oxide equivalent being from .20 to .33 of said aluminum nitrate, the amount of said aluminum nitrate being such as to yield 13.95 grams of aluminum oxide, and the amount of said magnesium nitrate being 47.25 grams.

3. Method of preparing a relatively stable insulating coating material containing aluminum nitrate, comprising heat treating commercial hydrolyzed aluminum nitrate to increase its aluminum oxide equivalent, mixing a relatively small portion of said coating material containing said heat treated aluminum nitrate, plating said coating material on a metal base as a test to determine the amount of said aluminum oxide equivalent, and repeating said heat treatment and said test until said test produces a relatively smooth coating, said relatively smooth coating indicating an aluminum oxide equivalent of from 20 to 33% by weight of the treated aluminum nitrate.



4. An electrophoretic insulating coating material comprising aluminum oxide, magnesium nitrate, aluminum nitrate and a solvent, the amount of said aluminum nitrate being determined by dividing a constant having a value of 13.95 by the aluminum oxide equivalent of said aluminum nitrate, said aluminum oxide equivalent having a range of from 20 to 33% of said aluminum nitrate by weight.

HENRY T. SWANSON.

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