

PATENT SPECIFICATION

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(54) METHODS OF COATING DISCHARGE LAMP ENVELOPES

(71) We, WESTINGHOUSE ELECTRIC CORPORATION, of Westinghouse Building, Gateway Center, Pittsburgh, Pennsylvania, United States of America, a company organised and existing under the laws of the Commonwealth of Pennsylvania, United States of America, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

This invention relates to methods of coating spectrum-modifying particles, such as phosphors or pigments, onto discharge lamp envelopes.

Discharge lamp envelopes are usually coated by suspending a finely divided material (such as phosphor) in a liquid vehicle. The liquid vehicle has dissolved therein an organic binder which imparts the proper viscosity and suspending properties to the vehicle. This suspension (or so called "paint") is flowed over the interior surface of the envelope, the excess drained, the liquid vehicle volatilized, and the coated tube heated (lehdred) to drive off the binder material. Typically the lehring is in an oxidizing atmosphere and the binder material is removed by oxidization. While organic solvents were widely used at one time, water vehicle systems are now generally used as such water systems are cheaper and safer. A variety of water soluble binders have been used, including methyl cellulose, polyacrylic acid, and some hydroxy alkyl celluloses. The most widely used system is probably hydroxy ethyl cellulose.

Hydroxyethyl cellulose is typically lehdred at about 650°C for approximately 60 seconds (the aforementioned water soluble binders are all generally lehdred under approximately these same conditions). As these binders are removed by pyrolysis and oxidation, the 650°C lehr temperature is needed to reduce the residual carbonaceous "char" to the

level of a few parts per million (failure to reduce the char to these levels results in brightness and lumen loss in the completed lamp). The requirement for high lehring temperature does present problems, however, as it discolours some pigments (such as calcium sulfide/selenide) and requires large amounts of fuel to heat the lehring furnace.

Prior art coatings systems have used finely divided particles such as alumina or silica as adhesion promoters. Such adhesion promoters are typically used in amounts of up to about 1/2 of 1% of the phosphor weight (it had generally been found that a greater amount provided no further improvement in adhesion and reduced lamp efficiency). In U.S. Patent 3,424,605 is noted the addition of finely divided silica to promote adhesion, with the comment that it can be deleterious to lamp efficiency.

The aforementioned United States patent also discloses the use of polyethylene oxide (sold under the name "Polyox" by Union Carbide, "Polyox" is a registered Trade Mark, as a binder for lamp coating, the adhesion problem with polyethylene oxide and the addition of Ba(NO₃)₂. The temporary binder is lehdred out at 600—635°C and then the temperature is raised to 670°—810°C to melt the Ba(NO₃)₂ to obtain adequate adhesion.

According to the present invention a method of coating a discharge lamp envelope comprises preparing a coating composition comprising spectrum-modifying particles, alumina particles, and an aqueous solution of polyethylene oxide having a viscosity of from 20—200 centipoises, said alumina particles being present in an amount such that the weight ratio of alumina particles to spectrum-modifying particles is rfrom 0.01—0.04; coating said discharge lamp envelope with said coating composition; and lehring said coated envelope at a temperature of from 400—550°C.

This method provides significant savings in fuel for the lehring furnaces, the possibility of coating materials which are unstable at higher lehring temperatures, and the possibility of coating particles which are subject to oxidation in air at lehring temperatures (as the lehring can be performed in a non-oxidizing atmosphere).

It has been discovered that satisfactory adhesion can be obtained using a polyethylene oxide binder only if a relatively large amount of very fine aluminium oxide (greater than 1% of the phosphor weight) is included in the coating composition. This result is surprising as, with prior binder systems, the use of greater than 0.5% of alumina had apparently never further improved adhesion. It was further discovered that lamps using the polyethylene and high alumina combination produced lumen output as good if not better than lamps using a hydroxyethyl cellulose binder containing about 0.5% alumina. This result was also surprising as high alumina content in prior art lamps had resulted in relatively low efficiency.

Although the mechanism is not completely understood, it is felt that a unique synergism occurs in this system that allows the use of relatively large amounts of finely divided alumina whereas such amounts are deleterious to lamp performance in a normal binder system. This synergism arises because the rapid rate of gas evolution in the pyrolysis of polyethylene oxide causes a very "loose" or non-compact structure. The open nature of the powder layer apparently allows much greater amounts of alumina to be added without significantly increasing the scattering light loss that normally prevents such high "load-up" of alumina.

It is considered therefore that the combination of a rapid pyrolysis binder as polyethylene oxide represents a unique situation can profitably utilize relatively large amounts of alumina.

Polyethylene oxide also has the advantage (compared to hydroxyethyl cellulose) of being resistant to attack by micro-organisms. The viscosity of the coating composition must be accurately controlled both to maintain the particles in suspension and to provide for a proper coating thickness on the lamp envelope. If, as is common in production processes, the binder solution is stored for some period of time, micro-organisms will attack a binder such as hydroxyethyl cellulose and cause a dramatic loss of viscosity (if special precautions are not taken). Polyethylene oxide, on the other hand, is micro-biologically stable and thus the difficulties from possible micro-organisms attack are avoided.

The most important advantage of the polyethylene oxide and high alumina system is probably the reduction in lehring temperature. Measurements have indicated that the natural gas fired lehring furnaces require 30% less of natural gas when the temperature is lowered from about 650°C. to about 500°C. This provides a very significant saving in fuel cost. The lower temperature also allows usage of a much wider range of phosphors. Copper activated phosphors and sulfide phosphors have generally been rejected as unusable because of the 650°C lehring, but can be used if lehring at about 500°C. A wider range of pigments can also be used. The 500°C lehring is also convenient in that it is sufficient to relieve any residual stress from the drawing and cutting of the lamp envelopes, but provides less stress cracking than does 650° lehring. The lower lehring temperature also minimizes the reaction of various phosphors with sodium in the glass. Zinc silicate phosphors, for example, are sensitive to sodium darkening and are therefore benefited by a lower lehring temperature.

Polyethylene oxide is thought to break down to ethylene oxide gas upon heating to lehring temperature. The ethylene oxide gas then escapes from the coating with minimal char formation. This is in contrast to the prior art binders which form a carbonaceous char in the oxidation/pyrolysis which must be then further oxidized to remove the residual carbon (the lamp output being seriously affected if any of this residual carbon is not removed). The bakeout of polyethylene oxide does, however, result in a rapid gas evolution which can have the unfortunate consequence of disruption of the phosphor layer resulting in extremely poor phosphor adherence. A variety of adhesion promoters were investigated with varying amounts of each and only the relatively large percentage of finely divided alumina provided satisfactory adhesion. Among the adhesion promoters which were found unsatisfactory are calcium carbonate, barium oxide, $\text{Ba}(\text{NO}_3)_2$ (which was not raised to its melting temperature as taught by the aforementioned U.S. Patent 3,424,605), magnesium oxide, calcium nitrate, magnesium phosphate, barium tungstate, barium phosphate, $(\text{NH}_4)_2\text{HPO}_4$ and $\text{HBO}_3(\text{B}_2\text{O}_3)$. The amount of these adhesion promoters was generally varied between 0.5 and 2.0%. The use of only about 0.5% of finely divided formed alumina did not provide adequate adhesion. At higher finely divided alumina levels (about 1% of phosphor weight) major adhesion improvements occurred. At 1.5% the phosphor adhesion was comparable to the present hydroxyethyl cellulose system

(with its approximately 0.5% alumina) and at 2% alumina with polyethylene oxide there was excellent adherence (better than now obtainable with hydroxyethyl cellulose).

Fumed alumina with an approximately 300 Angstrom particle size (sold commercially as "Alon C" by Degussa of West Germany) has been found to be especially effective. (Alon[®] is a registered Trade Mark). It should be noted that precipitated alumina (with an approximately 600 Angstrom particle size) has been found to be significantly less effective. Whether the degree of effectiveness is dependent upon particle size or is dependent upon the method of preparation is not known at this time.

It has also been found that good texture is best provided by dispersing the alumina in water before mixing into the paint. It has also been found that better performance is provided when the flexibilizers normally used in lamp coating (such as triethanolamine or glycerol) are not used.

Tests have indicated that the major portion of the polyethylene oxide can be removed by lehring in air at 400°C, but that for rapid and reasonably complete removal, the lehring should be performed at about 500°C. The removal of polyethylene oxide in an inert atmosphere such as nitrogen does not occur to any significant extent at 400°C but is almost completely removed at about 450°C. In no case is lehring above 550°C necessary and, as it results in extremely rapid gas evolution, lehring at these higher temperatures is undesirable.

The weight ratio of alumina particles to phosphor (or other spectrum modifying particles) is preferably between 0.15 and 0.025. While weight ratios as low as 0.01 can be used, the adhesion in this range is somewhat less and some loss of the phosphor coating can occur when the completed lamps are handled. Alumina weight of 0.025 to 0.04 can also be used, but such loading is generally unnecessary for adhesion purposes.

Because polyethylene oxide does not require oxidation during lehring, it can be lehrd in a non-oxidizing atmosphere. This permits coating of a number of spectrum modifying particles which are subject to oxidizing in air.

With the exception of the flexibilizers, the commonly used additives such as wetting agents and the foaming agents are compatible with the polyethylene oxide system. For example, Bubble Breaker B8748 (Balab Inc.) can be used as a defoaming agent and Igepal C0610 (GAF) can be used as the wetting agent. (IGEPAI is a Registered Trade Mark).

The invention will now be illustrated by the following Examples:

EXAMPLE I.

In order to coat a cool white halophosphate phosphor, a stock solution of 8.0 grams of polyethylene oxide is dissolved in 1 liter of deionized water. The dissolved solution is then passed through a mixed bed polystyrene ion exchanger system in order to remove the impurities from the solution. About 1.4 gram of fumed aluminum oxide is dispersed in approximately 10 cc's of deionized water and this dispersion is added to about 100 cc's of the purified polyethylene oxide solution. Approximately 0.04 grams of wetting agent and 0.04 grams of defoaming agent is then added and then 70 grams of cool white halophosphate phosphor is stirred into the foregoing to form the paint. The resulting paint is then flowed over a fluorescent lamp envelope in the usual manner to provide such a thickness that about 7 grams of phosphor is deposited on a 40W T12 bulb. After coating, the envelope is dried to remove the water and then lehrd by heating to a temperature of approximately 500°C for about 1 minute in order to completely volatilize the residual polyethylene oxide binder metal.

In the foregoing paint, the amount of fumed aluminium oxide can be varied between approximately 0.7 grams and 2.8 grams, and is preferably between 1.05—1.75 grams. The wetting agent can be added in amounts of up to 1 gram and the defoamer can be added in amounts up to about 0.5 grams. The amount of phosphor can also be varied between 10 and 100 grams (thus providing one method of varying the thickness of the phosphor coating on the lamp envelope). Viscosities of about 20—200 centipoises are generally desired for coating paints. As known in the art, the viscosity can be adjusted by varying either the amount or molecular weight of the binder.

Example II.

In order to coat calcium tungstate phosphor, 1.0 grams of polyethylene oxide is completely dissolved in 100 cc's of deionized water. Approximately 1.75 grams of fumed alumina is dispersed in approximately 5 cc's of deionized water and added to the polyethylene oxide solution. Approximately 70 grams of finely divided calcium tungstate phosphor is then suspended therein. The coating composition is then coated on the discharge lamp envelope in the usual manner. The coated envelope is then lehrd for approximately 1 minute and about 450—550°C.

Example III.

In order to coat a pigment onto a fluorescent lamp envelope, 0.44 gram of

polyethylene oxide is dissolved in 55 cc's of deionized water. About 0.4 grams of finely divided alumina is dispersed in 44 cc of deionized water which is then mixed with the polyethylene oxide solution. Approximately 20 grams of cadmium sulfide/selenide pigment and 0.04 gram of wetting agent are then added. The pigment suspension is then coated on a discharge lamp envelope in the normal manner and the coated envelope is lehrred at a temperature of approximately 500°C.

As illustrated by the foregoing Examples, polyethylene oxide binder coating solutions containing finely divided alumina (in amounts of greater than 1% of the phosphor weight) can be coated in the normal manner and lehrred at less than 550°C. The flow properties of this coating composition are as good or better than hydroxy ethyl cellulose (and generally somewhat better than hydroxy propyl methyl cellulose) with the lower molecular weight varieties of polyethylene oxide having slightly better flow properties than the somewhat higher molecular weight polyethylene oxide paints. The viscosity of the coated composition can be adjusted by the one skilled in the art in the normal manner, with the lower molecular weight polyethylene oxide requiring the slightly higher number of grams per liter of water to provide a desired viscosity. It has been found that molecular weights in the range of about 300,000 to 400,000 provide very good performance.

WHAT WE CLAIM IS:—

1. A method of coating a discharge lamp envelope which comprises preparing a coating composition comprising spectrum-modifying particles, alumina particles, and an aqueous solution of polyethylene oxide having a viscosity of from 20—200 centipoises, said alumina particles being present in an amount such that the weight ratio of alumina particles to spectrum-modifying particles is from 0.01—0.04; coating said discharge lamp envelope with said coating composition; and lehring said coated envelope at a temperature of from 400—500°C.

2. A method according to claim 1, wherein the weight ratio of alumina particles to spectrum-modifying particles is between 0.015 and 0.025.

3. A method according to claim 1 or 2, wherein the lehring temperature is between 450°C and 550°C.

4. A method according to claim 1, 2 or 3, wherein the spectrum-modifying particles are subject to oxidation in air at lehring temperatures and said lehring is performed in a non-oxidizing atmosphere.

5. A method of coating discharge lamp envelopes as claimed in claim 1 and substantially as described herein with particular reference to the foregoing Examples.

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polyethylene oxide is dissolved in 55 cc's of deionized water. About 0.4 grams of finely divided alumina is dispersed in 44 cc of deionized water which is then mixed with the polyethylene oxide solution. Approximately 20 grams of cadmium sulfide/selenide pigment and 0.04 gram of wetting agent are then added. The pigment suspension is then coated on a discharge lamp envelope in the normal manner and the coated envelope is lehrd at a temperature of approximately 500°C.

As illustrated by the foregoing Examples, polyethylene oxide binder coating solutions containing finely divided alumina (in amounts of greater than 1% of the phosphor weight) can be coated in the normal manner and lehrd at less than 550°C. The flow properties of this coating composition are as good or better than hydroxy ethyl cellulose (and generally somewhat better than hydroxy propyl methyl cellulose) with the lower molecular weight varieties of polyethylene oxide having slightly better flow properties than the somewhat higher molecular weight polyethylene oxide paints. The viscosity of the coated composition can be adjusted by the one skilled in the art in the normal manner, with the lower molecular weight polyethylene oxide requiring the slightly higher number of grams per liter of water to provide a desired viscosity. It has been found that molecular weights in the range of about 300,000 to 400,000 provide very good performance.

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2. A method according to claim 1, wherein the weight ratio of alumina particles to spectrum-modifying particles is between 0.015 and 0.025.

3. A method according to claim 1 or 2, wherein the lehring temperature is between 450°C and 550°C.

4. A method according to claim 1, 2 or 3, wherein the spectrum-modifying particles are subject to oxidation in air at lehring temperatures and said lehring is performed in a non-oxidizing atmosphere.

5. A method of coating discharge lamp envelopes as claimed in claim 1 and substantially as described herein with particular reference to the foregoing Examples.

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