

# UNITED STATES PATENT OFFICE

2,325,408

## COATED GLASSINE PAPER

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No Drawing. Application August 5, 1942,  
Serial No. 453,720

9 Claims. (Cl. 117-86)

This invention relates to the coating of glassine paper and particularly to the coating thereof with waxy compositions.

The coating of glassine paper with wax-containing plastic materials has long been carried out by dissolving the wax-containing plastic material in a volatile solvent, applying the resulting lacquer to the paper and then evaporating the solvent. Although this method gives coatings which are satisfactory for many purposes, it is apparent that the method entails considerable expense since the large amount of solvent involved is not easily recovered and constitutes a considerable fire hazard and, furthermore, the time required for evaporation of the solvent renders the process slow and cumbersome.

To overcome these difficulties attempts have been made to coat glassine paper with the so-called ethyl cellulose "melts." These are non-solvent-containing ethyl cellulose compositions which become sufficiently fluid at elevated temperatures to permit their application directly to paper surfaces without the use of a solvent. Upon cooling, the melt solidifies to give a smooth, even coating. By the use of melts of this type, not only is the solvent loss eliminated, but the coating process may be carried out rapidly since the hot coated sheet of paper cools almost instantaneously after it leaves the coating machine. Various belt compositions of the type mentioned have been used in the coating of kraft, sulfite and similar relatively porous papers.

There are, however, certain difficulties inherent in this method of procedure as applied to glassine paper which it has not heretofore been possible to overcome. Non-waxy high viscosity melts yield coatings of good adherence on glassine paper and sheets of the so-coated paper may be readily sealed together to form joints and seams by means of heat and pressure. However, due to the high viscosity of such melts it is not possible to apply a desirably thin layer with the conventional coating machines used in the industry, such as the Waldron and Mayer machines, and the resulting thickly coated paper is undesirably stiff for many purposes. Coating costs are correspondingly high due to the material consumed in forming the thick layer. Furthermore, these high viscosity melts soften only at relatively high temperatures, and the heating of the glassine sheet to the temperature required for the coating with, or the sealing of, these high viscosity melts leads to considerable dehydration of the fibers and embrittlement of the glassine sheet.

Although melts which soften at lower tem-

peratures and which have lower viscosities have been used successfully to coat sulfite, kraft, and other porous papers which they penetrate readily, they do not adhere well to the highly hydrated fibers in glassine paper because they invariably contain waxes to lower the viscosity and softening point. Such coatings are likely to separate from the paper during use. Separation of the coating from the sheet is particularly pronounced when the glassine sheet is of the plasticized type. Sheets of glassine paper coated with these low viscosity melts by previously known processes may be sealed together to form seams and joints by the application of heat and pressure, without appreciable embrittlement of the sheet because of the low sealing temperature required, but the application of tension to the seal or joint invariably leads to separation of the coating from the paper in the region of the joint. Melts having a viscosity of about 1300 centipoises or higher as determined in the Ubbelohde viscosimeter at 105° C., have heretofore been used in order to secure good adhesion to the glassine and as stated above, the use of these high-viscosity melts causes considerable embrittlement of the glassine paper.

Coated glassine sheets have been prepared by applying an under-coating of an ethyl cellulose composition dissolved in a suitable solvent followed by evaporation of the solvent and subsequent application of the desired low viscosity ethyl cellulose melt. Although such methods yield tightly adherent coatings there is little if any decrease in the fire hazard or in the amount of volatile solvent lost since, to secure a satisfactory under-coating, almost as much solvent has to be used as is used in applying the complete coating in the form of a more concentrated solution of the solid coating ingredients. There is also little, if any, saving in drying time over the use of a conventional solvent coating.

It is, therefore, an object of the present invention to provide a method whereby thin, adherent coatings of low viscosity ethyl cellulose melts may be applied to glassine paper.

An additional object is to provide a glassine paper coated with a thin, adherent coating of a low viscosity ethyl cellulose melt.

These and related objects are readily accomplished by first applying to the glassine paper a thin under-coating of an aqueous emulsion of an ethyl cellulose lacquer and, after drying the paper, applying an over-coating of a low viscosity ethyl cellulose melt.

This new method possesses several advantages over the hitherto available methods both as re-

gards the economy and ease of carrying out the operation and, also, as regards the desirable properties of the coated glassine sheet prepared. Thus, the use of an aqueous emulsion of the ethyl cellulose lacquer for depositing the under-coating permits the use of a greatly reduced amount of volatile organic solvent without reducing the degree of penetration of the under-coating composition into the sheet, since the aqueous emulsion penetrates the sheet readily. Upon drying the sheet, there remains a thin, firmly bonded under-coating on the surface of the sheet comparable to that obtained when using a wholly organic solvent solution of the under-coating composition. Furthermore, the coated sheet may be dried rapidly and without danger of embrittlement since the presence of the water in the emulsion not only prevents dehydration of the fibers, but, also, during its evaporation carries with it the organic solvent used in making up the lacquer. Consequently, by the time the water content of the sheet has been lowered during the drying process to the point where the paper is sufficiently dry for use in the next step of the process but where there is still sufficient moisture left in the paper to prevent embrittlement, the organic solvent will all have been vaporized and the under-coating deposited in condition to receive the over-coating of low viscosity ethyl cellulose melt.

Although any of the usual lacquers of good film-forming qualities containing an organic solvent-soluble ethyl cellulose may be used in preparing the aqueous emulsion, lacquers containing a plasticizer for the ethyl cellulose, such as di(ortho-zenyl) monophenyl phosphate, dibutoxyethyl phthalate, tertiary butyl phenoxy ethanol, glycerol mono stearate, etc., are preferred, since the presence of such plasticizer renders the under-coating more flexible. It is also preferable to avoid the use of high boiling organic solvents in the lacquer since these tend to prolong the drying period following the application of the aqueous emulsion. The solvents, and mixtures thereof usually used in formulating ethyl cellulose lacquers include toluene, xylene, butanol, the so-called aromatic solvent fractions of petroleum, naphtha-like aromatic products obtained by the simultaneous cracking and hydrogenation of petroleum, etc. Such aromatic petroleum products are available commercially under a variety of trade names including "Union aromatic solvent," "Solvesso," "Solvasol," "Hydroformed solvents," etc. It is desirable to avoid the use of water-miscible solvents, such as ethanol, although small proportions of such solvents may be included if desired.

The aqueous lacquer emulsion may be prepared in any convenient manner and any emulsifier or wetting agent capable of emulsifying the lacquer may be used. Emulsifiers which have been used with advantage include the commercially available products "Aresklene," said to be a sodium disulfonate of dibutyl phenyl phenol, and "Santomerse No. 3," said to be a salt of a substituted aromatic sulfonic acid. The emulsifying agent may be dissolved in the desired amount of water and the lacquer then added with agitation. Although the amount of emulsifying agent used will depend upon the particular agent and the particular lacquer used, from 0.5 to 5.0 per cent solutions of the emulsifier in water have been used to advantage. Greater or lesser amounts may, however, be used provided the lacquer is dispersed sufficiently to deposit an even coating

of the solid ingredients on the surface of the glassine paper upon drying the sheet. Emulsions containing from 4 to 35 per cent, and preferably from 5 to 30 per cent, by weight of solids may be used and a satisfactory under-coating obtained with one application of the emulsion. Emulsions having a lower content of solids may, of course, be used, although it may be necessary to make more than one application of the emulsion. After the glassine sheet has been coated with the emulsion it may be dried in any convenient manner, such as by blowing warm air over it or by passing the sheet over heated rolls. Heating for prolonged periods at temperatures above about 100° C. should be avoided to prevent dehydration of the fibers and embrittlement of the glassine sheet. The degree of heating which may be applied to the sheet without embrittling it will be apparent to anyone skilled in the art.

Although the under-coating may be applied to the finished glassine sheet as previously described the invention is not limited to the application in such manner. If desired, the under-coating may be applied during the manufacture of the glassine paper on the paper machine. Thus a size press may be inserted in the drying roll train and the ethyl cellulose lacquer emulsion applied to the partially dried sheet by means of the press. The emulsion may also be applied to the dried sheet either before or after curing and before the supercalendering operation. It is to be understood that the term "pre-coating a glassine paper sheet" as used herein includes the forming of the emulsion-deposited coating on the sheet regardless of whether the operation is performed on the sheet at some stage during its formation or on the finished, supercalendered sheet.

The dried sheet is then coated with the low viscosity ethyl cellulose melt. Thin, flexible adherent over-coatings of the low viscosity ethyl cellulose melt may be applied with conventional coating machines, such as the Waldron or Mayer machines, at temperatures sufficiently low to avoid embrittlement of the glassine. Ethyl cellulose melts having viscosities of as low as 300 to 1000 centipoises, when determined in the Ubbelohde viscosimeter at 105° C., may be applied at temperatures insufficiently high to cause embrittlement of the glassine sheet and excellent adherence of the over-coating to the emulsion coated sheet obtained. The melt may also be applied in other ways which are apparent to anyone skilled in the art, the choice of the method of application depending largely upon the type and thickness of the coating desired.

Any ethyl cellulose melt of low viscosity may be used to form the over-coating. Waxes such as 12-hydroxy stearin, known commercially as "opal" wax, and paraffin wax, which are ordinarily incorporated in the melt to lower its viscosity and to reduce its vapor transmission do not prevent good adhesion of the melt to the emulsion coated sheet. The melt may contain resins compatible therewith to increase the toughness of the film and, also, plasticizers to increase the flexibility of the film. Suitable ethyl cellulose melts may include from 5 to 15 per cent by weight of an organo-soluble ethyl cellulose having a viscosity of from 10 to 20 centipoises when measured as a 5 per cent solution in a mixture of 80 per cent toluene and 20 per cent ethanol by the falling ball method as outlined in A. S. T. M. D-301-33, from 25 to 50 per cent of an ester wax, such as opal wax, 20 to 35 per cent of a mineral wax, such as paraffin wax, 5 to 20

per cent of a plasticizer, such as dibutyl phthalate, the non-drying pure alkyd resins, paraffin oil, methyl abietate, etc., and from 20 to 35 per cent of a compatible resin. Resins which have been used to advantage include pure phenolic resins, oil modified phenolic resins, alkyd resins and oil modified alkyd resins. It should be pointed out, however, that the invention is not limited by the type of wax, resin or plasticizer or by the proportion thereof used in the low-viscosity ethyl cellulose melt, or by the type or proportion of organo-soluble ethyl cellulose in the melt provided, however, that the viscosity of the melt is sufficiently low to permit its application to the emulsion coated glassine paper and heat sealing of the finally coated sheet at temperatures below those at which the sheet is embrittled.

Coated glassine paper prepared by the herein described method is strong, flexible, and waterproof. The coating is strongly bonded to the paper and cannot be separated from it without rupturing the paper. Sheets of the coated glassine paper may be sealed readily by the application of heat and pressure without decreasing the strength of the coated sheet in the region of the seal. The heat sealing qualities of the coated sheet are not appreciably altered upon aging.

Certain advantages of the invention will be apparent from the following examples which are not to be construed as limiting the invention.

#### Example 1

An ethyl cellulose having an ethoxy content of 49 per cent and a viscosity of 10 centipoises, when measured as a 5 per cent solution in a mixture of 80 per cent toluene and 20 per cent ethanol by the falling ball method as outlined in A. S. T. M. D-301-33, was used in preparing a lacquer having the composition in parts by weight:

	Parts
Ethyl cellulose.....	70
Di(ortho-xenyl) monophenyl phosphate.....	30
Solvesso No. 1.....	158
Butanol.....	28

Two parts by volume of the lacquer were emulsified by pouring it with agitation into 1 part of water containing 1 per cent of Santomerse No. 3. A thin coating of the emulsion was then spread onto a sheet of plasticized glassine paper and the sheet allowed to dry by exposure to air at the ordinary room temperature. The sheet was then coated with a thin layer of an ethyl cellulose melt having the composition in parts by weight:

	Parts
Ethyl cellulose (26 centipoise, 49 per cent ethoxy).....	10
Opal wax (hydrogenated castor oil).....	22
Paraffin wax (m. p. 135° F.).....	23
Di-butyl phthalate.....	10
Super-Beckacite 2000 (a pure phenolic resin).....	35

The melt was applied at a temperature of about 105° to 120° C. The coated glassine sheet was extremely flexible and the coating adhered tenaciously to the sheet under all conditions of flexing and tearing. No embrittlement of the sheet occurred during the coating process or during the sealing together of separate sheets of the coated paper by the application of heat and pressure. The seals produced by the application of heat and pressure were stronger than the rest of the

coated sheet and did not separate from the paper under stress. This example illustrates the improvement in heat seal strength accomplished by the use of ethyl cellulose under-coating over that obtained on untreated glassine paper. Seals formed in the same manner as the above but on untreated paper separate easily from the paper under stress.

#### Example 2

A glassine sheet was treated with an aqueous emulsion of an ethyl cellulose lacquer and dried as in Example 1. The sheet was then coated with a thin layer of an ethyl cellulose melt having a viscosity of about 450 centipoises at 105° C. and having the composition in parts by weight:

	Parts
Ethyl cellulose (10 centipoise, 49 per cent ethoxy).....	11
Opal wax (hydrogenated castor oil).....	30
Paraffin wax (m. p. 150° F.).....	23
Di-butyl phthalate.....	8
Amberol 800 P (maleic modified alkyd resin).....	20
Beckasol 24 (pure alkyd plasticizer).....	8

The coated glassine sheet was flexible and strong and the coating was firmly bonded to the sheet. No embrittlement of the sheet occurred and seals of ample strength were obtained with moderate heat and pressure.

We claim:

1. The method which includes: pre-coating a glassine paper sheet with an aqueous emulsion of an ethyl cellulose lacquer; volatilizing water and volatile solvents from the pre-coated sheet; and coating the dried sheet with a low viscosity ethyl cellulose melt.

2. The method which includes: pre-coating a glassine paper sheet with an aqueous emulsion of an ethyl cellulose lacquer; volatilizing water and volatile solvents from the pre-coated sheet; and coating the dried sheet with a low viscosity ethyl cellulose melt, said drying and coating steps being carried out at a temperature below the embrittling temperature of the glassine sheet.

3. The method which includes: pre-coating a glassine paper sheet with an aqueous emulsion of an ethyl cellulose lacquer; volatilizing water and volatile organic solvents from the pre-coated sheet; and coating the dried sheet with an ethyl cellulose melt having a viscosity of from 300 to 1000 centipoises at 105° C.

4. The method which includes: pre-coating a glassine paper sheet with an aqueous emulsion of an ethyl cellulose lacquer, said emulsion containing from 4 to 35 per cent by weight of solids; volatilizing water and volatile organic solvents from the pre-coated sheet; and coating the dried sheet with an ethyl cellulose melt having a viscosity of from 300 to 1000 centipoises at 105° C.

5. In a method for coating a glassine paper sheet with a low viscosity ethyl cellulose melt, the steps which include: pre-coating the sheet with an aqueous emulsion of an ethyl cellulose lacquer; and vaporizing water and volatile solvents from the pre-coated sheet.

6. In a method for coating a glassine paper sheet with a low viscosity ethyl cellulose melt, the steps which include: pre-coating the sheet with an aqueous emulsion of an ethyl cellulose lacquer containing from 4 to 35 per cent by weight of solids; and vaporizing water and volatile solvents from the pre-coated sheet.

7. In a method for coating a glassine paper sheet with an ethyl cellulose melt having a vis-

cosity of from 300 to 1000 centipoises at 105° C., the steps which include: pre-coating the sheet with an aqueous emulsion of an ethyl cellulose lacquer; and vaporizing water and volatile solvents from the pre-coated sheet.

8. As an article of manufacture, a glassine paper sheet coated with a low viscosity ethyl cellulose melt and having interposed between the sheet and the melt coating an intermediate coating formed by pre-coating the sheet with an aqueous emulsion of an ethyl cellulose lacquer and drying the pre-coated sheet.

9. As an article of manufacture, a glassine

paper sheet coated with an ethyl cellulose melt having a viscosity of from 300 to 1000 centipoises at 105° C., said coated sheet having interposed between the melt coating and the glassine sheet  
5 an intermediate coating formed by pre-coating the sheet with an aqueous emulsion of an ethyl cellulose lacquer and volatilizing water and volatile solvents from the pre-coated sheet, said emulsion containing from 4 to 35 per cent by  
10 weight of solids.

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