COATED PAPER AND METHOD OF PRODUCING THE SAME
Filed Mar. 26, 1962, Ser. No. 183,304
10 Claims. (Cl. 161—250)

The present invention relates to an improved method of producing coated paper and to the coated paper produced particularly by a method of producing polyolefin coated paper having heightened resistance, i.e., improved barrier properties, to the passage of gases such as water vapor and solvents such as fats and oils.

This is a continuation-in-part of our copending application Serial No. 135,884, filed January 4, 1961, now Patent No. 3,161,560.

In recent years paper coated with polymers and copolymers of the lower olefins have been widely used in the packaging of products, including both solids and liquids. Polyethylene has been the most widely used material for this purpose, and the invention will be described primarily in connection with polyethylene. However, the principles of the invention are applicable generally to the polymers and copolymers of the lower olefins, e.g., propylene, ethylene and butylene, which may be extruded in molten form and deposited on a moving paper web. The term “lower olefins” as used herein is intended to mean the polymers and copolymers of propylene, ethylene and butylene. The principles of the invention are also applicable to substrate materials other than paper which have dielectric constants similar to paper.

The coating of a polyolefin, such as polyethylene, upon paper is usually accomplished by passing the extruded polyethylene film and the paper simultaneously between two pressure rolls and thereby bonding the hot film thoroughly to the paper. One of the pressure rolls is usually rubber-covered and is shielded by the paper web. The other of the pressure rolls is conventionally metal-covered, e.g., chrome-plated and serves to secure the release of the hot extruded polyethylene film. In this regard, it has been found that the use, on the premise that its excellent release properties are well known, of a hot pressure roll having a silicone rubber covering in place of the high cooling capacity chrome-plating does not successfully prevent the sticking of the hot extruded polyethylene to such pressure roll and the resultant production-halting breaking of the paper web. It has also been found that, even at relatively low speeds, the polyethylene film that forms the paper coating must be chilled from extruding temperatures of from about 260° to 315° C. to near room temperatures by the chrome-plated roll in a fraction of a second in order to release therefrom. It appears, therefore, that shock cooling is a requisite of the successful coating of paper with polyolefins.

A brief analysis of the nature of polyolefins, using polyethylene as a typical example, is necessary to a proper understanding of what the problem is. Polyethylene molecules are either linear or side-chained and compositions containing them are readily identifiable by their molecular weights, molecular weight distributions, densities, per-
polymers with which one can coat paper. More particularly, it has been taken to lie in the direction of ascertaining any bad effects on the thermal history of such coating films of their being shock-chilled to enable their release from the pressure loss roll bonding and therefore overcome or at least mitigating such effects. So much then for the problem giving rise to and the objects satisfied by the present invention.

It has now been found that, in a continuous process of coating paper with a film of a compound selected from the group consisting of polymers and copolymers of lower olefins, the pressure rolls commonly employed to bond the film to paper can be eliminated, thereby reducing equipment purchase and maintenance costs, and that shock chilling to obtain film release from one of the pressure rolls can be eliminated, thereby affording the film a thermal history consonant with either an effort to obtain improved barrier properties for smaller amounts of film raw material or an effort to maintain at or improve to a point approaching the theoretical maximum barrier properties of a given amount of film material. Thus, in our copending application Serial No. 80,586, filed January 4, 1961, we have described and claimed a process for producing improved polyethylene film coated papers in which the hot extruded film is applied to the paper pneumatically.

In the method of the present invention, a highly concentrated electric field is created and used to apply the hot extruded film to the paper. The method of the present invention has been found to yield results at least equal to those of our aforementioned copending application and far superior to those of the conventional practices described above, particularly as regards obtaining a high moisture vapor barrier at low coating weights. Another important advantage of the present invention over the conventional practices and over the method of the aforementioned copending application has been in the ease of obtaining a good polyethylene-paper adhesion at low coating weights.

The principal object of the present invention has been to provide a novel and improved method of coating a substrate such as paper with a film of a polymer or copolymer of the lower olefins, such as polyethylene. Another object of the invention has been to provide such a method in which superior moisture vapor barrier characteristics are achieved. Still another object of the invention has been to provide such a method in which electrostatic fields are used to produce adhesion of the film to the substrate.

A further object of the invention has been to provide such a method which is especially adapted to high speed operation. Another object of the invention has been to provide a polyethylene coated paper having improved moisture vapor barrier characteristics as compared to a paper coated with the same thickness of polyethylene by prior processes. Other and further objects, features and advantages of the invention will appear more fully from the following description of the invention.

The invention will now be described in greater detail with reference to the appended drawings, in which:

FIG. 1 is a schematic diagram illustrating one form of apparatus for practicing the method of the invention; Fig. 2 is a schematic diagram illustrating the operation of FIG. 1; and FIGS. 3-12 are schematic diagrams each illustrating a modified form of apparatus for practicing the method of the invention.

Referring now to the drawings and more particularly to FIG. 1, a continuous web 20 of paper from an unwind stand or other suitable source passes over a roll 21. The roll 21 is provided to pre-coat a surface of the web 20 with an adhesion promoter or primer such as Adco Chemical Company's Shawnad 313 which is a water-based solution containing 20% solids. The adhesion promoter, diluted to contain 1.5% solids, is contained in a trough 22 and is applied to one surface of web 20 as the web contacts roll 21. The roll 21 is in turn provided with the adhesion promoter by contact with roll 23 which extends into the trough 22. As the web 20 advances, roll 21 rotates simultaneously and therefore overcomes or at least mitigating such effects. So much then for the problem giving rise to and the objects satisfied by the present invention.

The web 20 leaving roll 21 passes around steam heated rolls 24, 25, and 26, idler rolls 27 and 28, steam heated rolls 29 and 30, and is electrically grounded backing roll 31, an idler roll 32 and a cooling roll 33. The web leaving the cooling roll 33 goes on to a winding roll, although other treatment zones may be positioned between the cooling roll and the winder. The grounded roll 31 and the cooling roll 33 are preferably driven rolls and the stem heated rolls may be driven if desired.

An extruder 34 and die 35 are positioned to extrude a thin film 36 of polyethylene or other plastic downwardly so as to contact a surface of the paper web 20 as the paper is passing around the roll 31. If the paper has been precoated, the pre-coated face faces outwardly of the roll 31 so as to contact the film.

From roll 31 onwardly, the paper web 20 has adhered to one surface thereof a thin film of polyethylene. The conventional practice in the past has been to cause the polyethylene film to adhere to the paper surface by causing the paper and the film to pass between two pressure rolls, as described above. In the method of copending application referred to above, adhesion of the polyethylene film to the paper surface is produced by pneumatic means. In accordance with the method of the present invention, electrostatic charges are used to apply the hot extruded polyethylene film to the paper with sufficient pressure to produce adhesion of the film to the paper.

More particularly, air in the region of convergence of the polyethylene film and the paper is ionized, and the ions are caused to impinge on the polyethylene film. The ions carry a charge and when they impinge on the polyethylene film the latter becomes charged. Since the paper is essentially uncharged, or at least possesses a different charge, a charge differential will exist between the polyethylene film and the adjacent paper. The resultant electric field between the polyethylene film and the paper will cause the film to be attracted to the paper.

The attractive field between the polyethylene and the paper must exist in the region of a distance between the polyethylene and the paper and hence the source of ions must be located so that the polyethylene film in the region of convergence will be charged to create the attractive field.

The charge density on the polyethylene surface could be expressed in terms of coulombs, while the attractive force resulting from the charge differential could be considered as a field strength expressible in coulombs per square centimeter. The greater the strength of this field the greater will be the force with which the polyethylene film is urged into contact with the paper surface.

In FIG. 1 the source of ionization is shown as a series of six horizontal wires vertically spaced in a vertical plane. The wires, which are designated by the reference numeral 37 extend across the width of the polyethylene film with the top wire approximately at the height of the top of roll 31. The wires 37 might be, for example, 3 mil tungsten wires separated from each other by a distance of not less than about 1/4", and sufficiently held in a holder 38 under sufficient tension to resist the substantial pull which will be experienced in the direction of the grounded backing roll 31. The spacing between the wires 37 and the polyethylene film may be of the order of 44" to 2", depending on the film thickness and the voltage applied to the wires. The current supplied is equally divided to the various wires 37, depending on electrical field configuration may be of the order of 3,000 to 50,000 volts, and may be positive, negative or alter-
nating. It has, however, been found more desirable to use a negative voltage on the wires. The voltage source is designated by the block 39 in FIG. 1 and may be of any suitable type.

The high voltage on the wires 37 ionizes the air in the immediate vicinity of the wires, and under appropriate conditions this can be observed as a corona discharge in the immediate vicinity of the wires. The corona should be limited to the region of the wires and preferably should not extend to the polyethylene film. The ions migrate away from the vicinity of the wire and some of them impinge on the polyethylene, resulting in an attractive electrostatic force between the polyethylene film and the paper, as described above.

A substantial proportion of the ions produced in the vicinity of the wires 37 will tend to travel in directions in which they will not impinge on the polyethylene film or at least not in the region of convergence between the polyethylene film and the paper. Various means can be used to concentrate the ion flow so as to maximize the ion flow into the polyethylene film in the region of convergence thereof with the paper. For example, air flow may be used to direct the ions in the desired direction. Or charged shields may be provided to inhibit ion flow except in the desired direction.

A physical explanation for the method of the invention is believed to be accurate but even if it should prove not to be accurate or to be incomplete, nevertheless the method of the invention can be practiced to advantage as herein described and claimed.

In the arrangement of FIG. 1 the diameter of the backing roll 31 is preferably relatively small, e.g., about 2 inches. The roll surface may be steel or may have a dielectric coating such as nylon, Hypalon, or fluorocarbon polyethylene sold by E. I. du Pont de Nemours & Co., Mylar (a polymerized ethylene glycol terphthalate), or Teflon (a tetrafluorethylene plastic). The dielectric coating is particularly desirable with small diameter backing rolls such as the 2″ diameter referred to, in order to minimize any tendency for arcing or sparking across the high voltage gap between the wires and the backing roll.

The polyethylene film 36 leaving the die 35 will generally have a temperature of about 600° F. but will cool so as to contact the paper at a temperature in the range of about 450-475° F. The paper web which contacts the polyethylene is preferably heated by the steam rolls to minimize chilling of the polyethylene; typically the paper temperature might be of the order of 150° F. to 240° F. at the backing roll 31. The heating of the paper also promotes adhesion of the polyethylene film to the paper.

When using small diameter backing rolls, e.g., the 2″ roll suggested for FIG. 1, the force on the polyethylene will tend to cause the polyethylene film to wrap around the roll, i.e., to contact the paper at a point in advance of the expected point of tangency of the film and the roll. This effect is graphically illustrated in FIG. 2 where the film 36 is shown as having a bow 36′ above the expected point of tangency 31′. Because of the bow 36′, the polyethylene film 36 contacts the paper 20 at point 31″, which is in advance of the point 31′. The distance between points 31′ and 31″ may be termed the amount of “wrap” or extra length of contact between the paper and the film. In general, the higher the applied voltage on the discharge wires 37 the greater will be the wrap.

However, the usable applied voltage is limited by the requirement that arcing between the wires and the grounded backing roll be avoided. The amount of wrap obtained is also dependent on speed and will decrease as speed increases. For example, with a field strength of ordinary magnitude and using a 2″ diameter backing roll, visible wrap will substantially disappear at paper speeds of about 400′ per minute.

In applying the polyethylene film to the paper substrate, there are two major factors to be accommodated. One is to achieve good adhesion of the polyethylene to the paper, and the other is to achieve a good moisture vapor transfer resistance (hereinafter termed “M.V.T.R.”).

With respect to adhesion, for some applications of the product of the invention it is sufficient that the polyethylene film be tightly bonded to the paper substrate so that delamination will not occur. Such an application might be as an inner wall of a multi-wall bag construction. But for other applications, where appearance of the product is important, it is desirable that the adhesion be uniform so as to avoid a patterned appearance. A patterned appearance may be in the form of visible lines in the polyethylene surface, areas of dull appearance, or in general regions whose light reflectance differs from the remaining surface. The presence of such regions seems to have little or no role on the film face are associated with the patterned effect and probably produce this effect, or at least vary the wrapping force on the polyethylene which in turn produces the pattern. The use of point sources instead of wires tends to produce a severe line patterned effect unless the point sources are located very close together, as in a fine wire brush.

The patterned effect has been found to be greater as the density of the paper substrate is increased, and it appears that to avoid a patterned effect on the product it is best to use low to medium density papers as the substrate. In general, it has been found difficult to avoid excessive patterned effect with a paper substrate density greater than about 75 seconds per 100 cc. measured by official standard T460 m-49 (April 1949) of the Technical Association of the Pulp and Paper Industry.

To obtain a satisfactory appearance, it is desirable that the paper substrate be uncoated. Thus the method of the invention, so far as it is presently known, is best applicable to coating polyethylene on a low to medium density uncoated paper.

The numerical M.V.T.R. figures set forth below are in terms of grams of moisture vapor transferred per 100 square inches of sample per 24 hours, after extrusion and lamination. The testing was in accordance with recognized testing procedure using a General Foods cabinet held at 100° F. and from 90 to 95% relative humidity.

Some specific examples of the method of the invention will now be given. The apparatus used was that of FIG. 1, using a 2″ diameter grounded backing roll and six 3 mil tungsten wires spaced ½″ apart in a vertical plane and spaced ½ inch from a vertical plane tangent to the adjacent side of the backing roll. The backing roll had a ½″ Hypalon covering. The voltage applied to the electrode wires was a negative 30,000 volts, producing an input current flow (at 110 volts) of 0.55 amps. The paper was a 40 lb. unbleached Kraft paper heated to about 175° F. The polyethylene was a high density Bake-Cole #7501.

The results of four tests (three specimens each) are tabulated in Table I. In Table I and in the other tables set forth, paper weight is in pounds per ream, i.e., pounds per 3000 square feet. Coating weight is likewise set forth in terms of pounds per ream (3000 square feet).
Further examples of the invention are set forth below in Table II and III for various coating weights of polyethylene on a 40 lb. unbleached kraft paper substrate. Table II is for density polyethylene (.945–.950), while Table III is for low density polyethylene (.918–.925).

Trials Nos. 1, 2, 3 and 4 of Table II are the same as the trials of Table I. The method of application for all the trials was the same as that described for Table I except for trials 9, 10, 11, 12, 17, 18, 19 and 20, which differed in that three tungsten wires were used in place of six, as described below in connection with FIG. 3. The "standard method" referred to is that employing pressure rolling and shock chilling. The coating weights and thicknesses set forth for the standard method are those which would be required to achieve the same M.V.T.R. reading as that shown in the tables for the corresponding trial using the method of the invention. The pounds or mls saved columns represent the saving in polyethylene coating weight or thickness by using the method of the invention rather than the standard method.

### TABLE II

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<th>Trial number</th>
<th>Coat wt. lbs.</th>
<th>Thickness mils</th>
<th>MVTR</th>
<th>Lbs. or mls saved</th>
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Average percent: 47

### TABLE III

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<th>Trial number</th>
<th>Coat wt. lbs.</th>
<th>Thickness mils</th>
<th>MVTR</th>
<th>Lbs. or mls saved</th>
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</table>

Average percent: 23
FIG. 3 shows a modified electrode structure for carrying out the method of the invention. FIG. 3 is similar to FIG. 1 but shows three parallel wires 37 in a holder 38 and arranged in a vertical plane in place of the six wires shown in FIG. 1. In FIG. 3, the wires are preferably spaced about ¼" apart. The backing roll 31 of FIG. 3 might be, for example, a ¼" nylon covered steel roll, and the roll-wire spacing might be ¼". The electrode wires are preferably tungsten, but might be made of other material, e.g., Nichrome wire. One or two wires could be used in place of the three wires of FIG. 3 or the six wires of FIG. 1, but at least three wires are preferred.

FIG. 4 is similar to FIGS. 1 and 3 except that the electrode wires are disposed in a curved surface concentric with roller 31. When six wires are used, the spacing between adjacent wires is preferably about ¼", but for three wires the spacing is preferably about ¼". The roll 31 in FIG. 4 might be identical with the roll shown in FIG. 3.

FIG. 5 shows the electrode structure as six parallel wires disposed in a horizontal plane with a spacing of about ¼" between adjacent wires. The vertical position of the electrode wires is preferably at the point of convergence of the polyethylene film and the paper, but satisfactory results have been achieved with the electrode structure moved somewhat up or down from that position.

FIGS. 6 and 7 show the electrode wires disposed in planes tilted 30° above and 30° below the horizontal, respectively.

FIG. 8 shows an arrangement in which the electrode wires are divided into two groups of three, with each group lying in a respective vertical plane. These planes might be, for example, ¼" apart.

FIG. 9 shows an electrode structure in which the wires 37 are replaced with two parallel bands 37' of 0.003" brass shim stock ¼" wide and spaced about ¼" from backing roll 31. A single band 37' has also been used successfully. The bands 37' have been successfully replaced with one or two ¼" wide bands of copper screen wire (200 mesh).

FIG. 10 illustrates the use of a large diameter backing roll. In FIG. 10, the backing roll 31 might be, for example, an 18" diameter chill roll having high gloss or matte finish. In FIG. 10 the electrode wires 37 are arranged in a vertical plane, as in FIG. 1, with the uppermost horizontal wire approximately at the level of the point of convergence of the polyethylene film 36 and the paper 20. An electrode structure of the type shown in FIG. 7 can be used to advantage with a large diameter backing roll.

FIG. 11 shows the use of a large diameter backing roll, as in FIG. 10, but the electrode 37A mounted in holder 38 is formed from a tube having an axially extending slit cut out of the side facing the backing roll. The tube 37A might be, for example, a 3/8" diameter copper tube and the slit might be ¾" wide. The slit should extend for the full width of the electrode.

In FIG. 12 the electrode 37B is formed by a series of razor blades having their sharp edges facing the backing roll. The razor blades should be mounted so that little or no gap exists between the polyethylene and the adjacent blades, or a single elongated sharp edged blade should be used in place thereof. As in the case of the electrode structures described, the electrode structure of FIG. 12 should extend across the complete width of the polyethylene and the holder should provide a firm mounting so that the electrode elements are held rigidly in position. The holders may be made of strong plastic or other suitable insulating material.

Satisfactory results have been achieved also using a 4" diameter ground steel backing roll with a 3/8" thick Hypalon cover and a 1 mil thick aluminum foil cover over the Hypalon.

While the invention has been described in connection with specific embodiments thereof and in a specific use, various modifications thereof will occur to those skilled in the art without departing from the spirit and scope of the invention as set forth in the appended claims.

What is claimed is:

1. The continuous process of coating a paper substrate with a film of a compound selected from the group consisting of polymers and copolymers of lower olefins, comprising the steps of extruding the film, advancing said substrate through a selected region, directing said extruded film along a path in which said extruded film and said substrate converge in said region, and ionizing the air in said region thereby subjecting said extruded film to said region to the action of an electric field whereby said film is forced into intimate bonding contact with said substrate.

2. The continuous process of coating a paper substrate with a film of a compound selected from the group consisting of polymers and copolymers of lower olefins, comprising the steps of extruding the film, advancing said substrate over and in contact with a grounded conductive roller located in space adjacent and the point of extrusion, directing said extruded film along a path through a region of convergence with and into contact with said substrate while the latter is passing over said roller, and ionizing the air in said region of convergence whereby subjecting said film in said region of convergence thereof with said substrate to the action of an electric field whereby said film is forced into intimate bonding contact with said substrate.

3. The process of claim 2 in which said substrate is maintained under substantial longitudinal tension while passing over said roller whereby said substrate is restrained against motion in a direction away from said roller and toward said film.

4. The continuous process of coating a paper substrate with a film of a compound selected from the group consisting of polymers and copolymers of lower olefins, comprising the steps of extruding the film, advancing said substrate over and in contact with a grounded conductive roller located in space adjacent the point of extrusion, directing said extruded film along a path through a region of convergence with and into contact with said substrate while the latter is passing over said roller, and subjecting the air in the space adjacent said region of convergence of said extruded film and said substrate to a high voltage ionized field whereby electrically charged ions produced by said field contact said extruded film in said region of convergence and force said film into intimate bonding contact with said substrate.

5. The continuous process of coating a paper substrate with a film of a compound selected from the group consisting of polymers and copolymers of lower olefins, comprising the steps of extruding the film, advancing said substrate over and in contact with a grounded conductive roller located in space adjacent and the point of extrusion, directing said extruded film along a path through a region of convergence with and into contact with said substrate while the latter is passing over said roller, and applying a high voltage between an electrode structure and said roller, said electrode structure being disposed in space adjacent the region of convergence of said film and said substrate, said voltage being sufficiently high to ionize the air in said space adjacent said region of convergence whereby resulting ions impinge upon said extruded film in said region of convergence and force said extruded film into intimate bonding contact with said substrate.

6. The process of claim 5 in which said voltage is of the order of 3,000 to 50,000 volts and in which the spacing between said electrode structure and said roller is about 1/4 inch to 2 inches.

7. The continuous process of coating a paper web with a polyethylene film, comprising the steps of heating said web, passing said heated web over a horizontally
disposed grounded metal roller, extruding a polyethylene film in space above said roller whereby said film passes through a region of convergence with and contacts said paper while the latter is passing around said roller, maintaining said web under substantial tension whereby said paper tightly contacts said roller, and applying a high voltage between said roller and an electrode structure positioned adjacent the region of convergence of said film and said web whereby the air adjacent said electrode structure is ionized and ions impinge on said film, said voltage and the relative spacing of said film and said structure being selected so that the attractive force produced between said film and said paper web will force said film into intimate bonding contact with said web.

8. The process of claim 7 in which said attractive force is substantially uniform across the width of said film.

9. The continuous process of coating a paper web with a polyethylene film, comprising the steps of heating said web, passing said heated web over a horizontally disposed grounded metal roller, extruding a polyethylene film in space above said roller whereby said film passes through a region of convergence with and contacts said paper while the latter is passing around said roller, maintaining said web under substantial tension whereby said paper tightly contacts said roller, and creating an ionized electrical field in said region of convergence of sufficient strength to produce an electrical charge differential between said film and said paper web in the region of convergence of said film and said web whereby said film is forced into intimate bonding contact with said web.

10. A paper substrate coated with a film of a compound selected from the group consisting of polymers and copolymers of lower olefins produced by the process comprising the step of extruding the film and the step of subjecting said film to the action of an ionized electrical field of sufficient strength to create a substantial electrical charge differential between the film and the substrate.

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UNITED STATES PATENT OFFICE
CERTIFICATE OF CORRECTION

Patent No. 3,196,063 July 20, 1965

Leon J. Paquin et al.

It is hereby certified that error appears in the above numbered patent requiring correction and that the said Letters Patent should read as corrected below.

In the grant, lines 2 and 3, for "Glenn M. Violette, of Greenwich, New Jersey," read -- Glenn M. Violette, of Greenwich, New York, --; in the heading to the printed specification, line 5, for "Glenn M. Violette, Greenwich, N. J.," read -- Glenn M. Violette, Greenwich, N. Y., --; column 1, line 57, for "requise" read -- requisite --; column 4, line 29, after "of" insert -- the --; column 7, line 26, after "for" insert -- high --.

Signed and sealed this 28th day of December 1965.

(SEAL)

Attest:

ERNEST W. SWIDER
Attesting Officer

EDWARD J. BRENNER
Commissioner of Patents