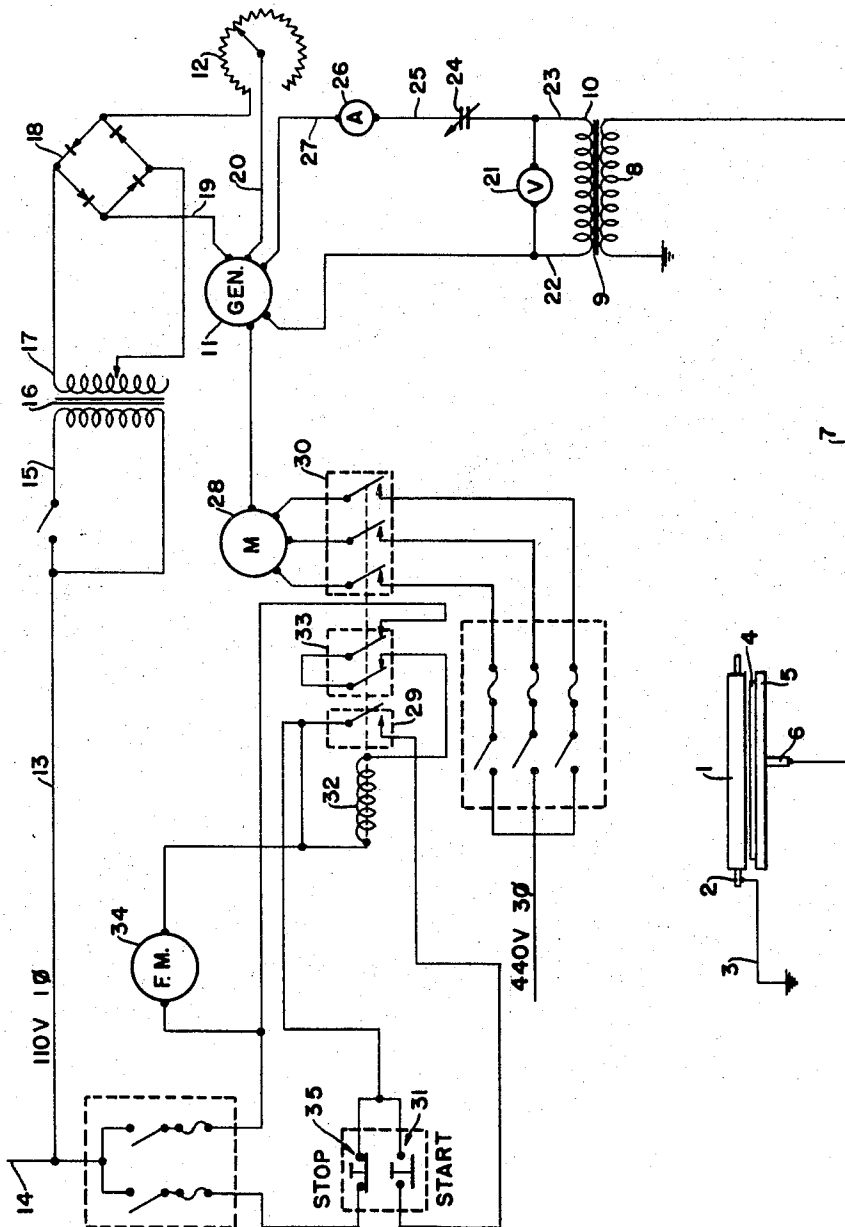


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ELECTRICAL DISCHARGE TREATMENT OF POLYETHYLENE



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**ELECTRICAL DISCHARGE TREATMENT OF
POLYETHYLENE**

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1 Claim. (Cl. 18—48)

This invention relates to the treatment of plastic material, and more particularly polyethylene, to improve the anchorage or adherence characteristics of the surface thereof. More particularly the invention is concerned with such a treatment or the control of such a treatment which does not destroy the heat sealability characteristic of the material or result in an unsatisfactory one.

In copending application Ser. No. 359,352, filed June 3, 1953, and assigned to the same assignee as is this invention, there is fully disclosed and described a method of treating polyethylene structures for the purpose of increasing the anchorage or adherence characteristics of their surfaces which comprises subjecting their surfaces to the action of a continuous electric glow discharge.

Copending application Ser. No. 474,904, filed December 13, 1954 and also assigned to the same assignee as is this invention, discloses and fully describes an improvement in the treating method of the prior application which involves subjecting a polyethylene structure to the electric glow discharge while at a temperature substantially above room temperature and advantageously at least at a temperature of 115° F. While the treatments of both of the aforementioned applications have proved satisfactory, some difficulty has been encountered in maintaining the heat sealability characteristics of the treated structures at a satisfactory level. The principal object of this invention is therefore to overcome such difficulty.

We have discovered that if, contrary to the prior practice of maintaining the current value of the electric glow discharge at a minimum dependent solely upon the basis of obtaining a satisfactory anchorage or adherence characteristic in order to consume a minimum of power, the current value of the glow discharge is raised to a point only sufficiently below the value at which arcing or burning through of the material to be treated would result to assure that no burning-through will occur, the heat sealability of the treated material will be greatly improved without degradation of the anchorage or adherence characteristics thereof. In other words, we have discovered that the anchorage characteristics of the surfaces of polyethylene structures can be improved in accordance with the treatments of said aforementioned applications and without destroying the heat sealability characteristics thereof if the current value of the electric glow discharge employed is maintained at a level only sufficiently below the burn-through value to assure an absence of burning or arcing-through. We have discovered that in most instances this current level is from 5% to about 15% below the current level at which a burning or arcing-through of the polyethylene structure would result.

While, as pointed out in the aforementioned copending applications, the electric glow discharge treatment with which we are here concerned is normally carried out by passing the material to be treated between an electrode and a grounded support for the material, in cer-

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tain instances it has been found desirable or necessary to employ a grounded support covered with a material having a high dielectric property. We have discovered further, in connection with the improvement of the present application, that heat sealability of the treated material is higher for substantially all current values of the electric glow discharge when the grounded support is so covered.

The above discoveries apply both to treatments carried out upon polyethylene at room temperature as well as upon polyethylene at temperatures substantially above room temperature.

In support of the above conclusions we have experimentally treated a large number of samples of polyethylene in sheet form both with the polyethylene at room temperature as well as with the polyethylene heated to substantially above room temperature. In the former case, an apparatus substantially similar to that disclosed in copending application Ser. No. 359,351, filed June 3, 1953 and assigned to the same assignee as is this invention was employed, the polyethylene film being taken from a supply roll thereof at room temperature. In the latter case, however, the polyethylene film was treated upon leaving the extruding apparatus by which it was formed. The details of treatment and the results achieved for a representative group of such experimental treatments are set forth hereinbelow. In all cases the frequency of the glow discharge, or perhaps more accurately of the discharge current in the electrode circuit, was about 2850 cycles per second. Except where indicated to the contrary, in all cases the grounded roll was bare, that is, uncovered.

In addition, in the following examples the tests for heat seal strength and degree of ink adhesion were the same in each case. In connection with heat seal strength the values given are in grams representing the force required to part sealed layers which were sealed at 240° F. under a sealing pressure of 30 pounds per square inch for 0.4 second. The values given for ink adhesion are the percentages of ink retained on the samples following a uniform ink stripping process; the "Dry" test being made following drying of the ink at room temperature usually for about 5 to 8 minutes; and the "Wet" test being made following immersion of the inked sample in crushed ice and water, storing it in a refrigerator for 24 hours, removing it therefrom and allowing the ice to melt, and wiping of the inked surface with tissue to remove loose ink and excess moisture. In neither case is it believed necessary to go into the details of the tests employed because the importance of the results is a relative one. Normally a treated film is considered commercially satisfactory from an ink adhesion point of view if, when subjected to the "Wet" test employed in connection with the following examples, a value of at least 60 to 80% is achieved.

While the current strength of the electrode circuit is referred to herein and in the claim as being that with the control of which this invention is involved, we have in fact in none of the following examples measured the electrical characteristics of the electrode circuit which is supplied from the secondary of a high voltage transformer, but rather have measured the electrical characteristics of the primary circuit of the transformer. This is because certain difficulties are encountered in measuring the characteristics of the secondary circuit which are not readily overcome. However, they need not be because the voltage-ampere characteristics of the electrode circuit correspond to those of the primary circuit; that is changes in the electrical characteristics of the primary circuit are reflected in direct proportion in the electrode circuit. Hence the voltage and current values given in the following examples are those for the primary circuit.

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As an aid to an understanding of the electrical setup employed, there is shown in the accompanying drawing a wiring diagram which will now be described.

A grounded roll 1 is connected through a metallic clip 2 and a ground wire 3 to ground. An electrode 4 from which a glow discharge is to be formed is supported by a micarta electrode holder 5 and is connected through a conductor rod 6 and line conductor 7 to one terminal of the secondary 8 of a high voltage transformer 9, the other terminal of which is grounded. The primary 10 of the transformer 9 (turn ratio is 1 turn on primary for each 40 turns on secondary) is provided with alternating current from a generator 11, the output of which may be varied by means of a field rheostat 12. A line conductor 13 from power source 14 connects to the primary side 15 of a powerstat 16 the secondary 17 of which is connected to a rectifier 18 which provides direct current to the generator 11 by line conductors 19 and 20. One terminal of the primary of the high voltage transformer 9 is connected to the generator through a conductor 23, a capacitor 24, a conductor 25, an ammeter 26 and a conductor 27. The other terminal is connected to the generator by a line conductor 22. A voltmeter 21 is connected across conductors 22 and 23. The insertion of the capacitor 24 in the primary circuit permits tuning thereof to reduce the voltage drop between the primary 10 of the transformer 9 and the generator 11.

The generator 11 is driven by a three-phase motor 28 connected to a 440 volt three phase source through normally open relay 30. The coil 32 of relays 29 and 30 is energized by pressing a start button 31 which closes relays 29 and 30 and opens the normally closed relay 33 across the lines of the power source. The closing of relay 29 effects the operation of the fan motor 34 to circulate air about the motor 28 and generator 11. Closing of the normally open relay 30 energizes the generator-driving motor 28. Relay 33 maintains a holding circuit through the coils of relays 29 and 30 so that the start button may return to inoperative position. In order to deenergize the circuits, stop button 35 may be depressed thereby deenergizing the coil of relay 29. This also deenergizes relay 30 whereby, when the stop button returns to its normally closed position, the system is not reactivated.

Example I

A polyethylene film made from a film-forming grade of polyethylene sold under the name of DYNH-3 by the Bakelite Corporation was treated substantially immediately following its withdrawal from the forming (extrusion) apparatus. Successive lengths were treated under the successive electrical conditions for the electrode circuit shown in the table below and were then subjected to heat seal strength and ink adhesion tests, the results of which are also shown in the table.

Primary Voltage in Volts	Primary current in milliamps	Heat Seal Strength (Gms.)	Ink Adhesion, Percent Retention	
			Dry	Wet
Untreated.....	Untreated.....	about 1,200..	0-5	0-5
155.....	420.....	110.....	50	50
160.....	610.....	40.....	95	95
163.....	800.....	90.....	100	100
168.....	1,000.....	150.....	95	85
169.....	1,100.....	185.....	100	85
170.....	1,220.....	Burn through		

Example II

Again successive samples of polyethylene film made from the same grade of film-forming polyethylene as the film of Example I, but from a different lot, were treated under the conditions and with the results set forth in the following table substantially immediately follow-

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ing the withdrawal of the film from the forming apparatus.

Primary Voltage in Volts	Primary current in milliamps	Heat Seal Strength (Gms.)	Ink Adhesion, Percent Retention	
			Dry	Wet
Untreated.....	Untreated.....	about 1,200..	0-5	0-5
152.....	440.....	95.....	100	95
152.....	520.....	75.....	100	95
156.....	600.....	75.....	100	95
160.....	680.....	190.....	100	95
162.....	810.....	495.....	100	95
163.....	940.....	620.....	100	95
163.....	1,000.....	Burn through		

Example III

In this experiment, successive samples of polyethylene film made from the same type of film forming polyethylene as employed in the prior examples but from different lots were treated, one series substantially immediately following formation of the film in an extrusion apparatus and the other on successive samples taken from a supply of film formed from the same lot of polyethylene by the extrusion apparatus but after they had cooled to room temperature.

TREATMENT AFTER EXTRUSION

Primary Voltage in Volts	Primary current in milliamps	Heat Seal Strength (Gms.)	Ink Adhesion, Percent Retention	
			Dry	Wet
Untreated.....	Untreated.....	680.....	0	0
148.....	300.....	100.....	90	50
151.....	390.....	110.....	100	80
157.....	480.....	45.....	100	90
161.....	620.....	165.....	100	90
160.....	700.....	350.....	100	80
162.....	900.....	455.....	100	50
169.....	1,200.....	Burn through		

TREATED AT ROOM TEMPERATURE

Untreated.....	Untreated.....	about 1,600..	0-5	0-5
160.....	290.....	1,545.....	20	20
160.....	350.....	1,110.....	30	50
161.....	440.....	705.....	50	50
166.....	700.....	675.....	95	90
171.....	1,200.....	Burn through		

Example IV

A further series of samples from the same grade of polyethylene, but still another lot, were treated under the following conditions and with the following results substantially immediately following formation in an extrusion apparatus.

Primary Voltage in Volts	Primary Current in Milliamps	Heat Seal Strength (Gms.)	Ink Adhesion, Percent Retention	
			Dry	Wet
Untreated.....	Untreated.....	970.....	0	0
235.....	260.....	300.....	50	50
239.....	520.....	60.....	100	90
242.....	710.....	30.....	100	80
245.....	950.....	1,600.....	100	60
248.....	1,200.....	Burn through		

Example V

A different brand of a film forming polyethylene was employed, namely, that designated Alathon 21-X manufactured by E. I. DuPont de Nemours was employed for comparison. A series of samples of this polyethylene were treated under the following conditions substantially

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immediately following formation in an extrusion apparatus and tested to obtain the following results:

Primary Voltage in Volts	Primary Current in Milliamps	Heat Seal Strength (Gms.)	Ink Adhesion, Percent Retention	
			Dry	Wet
Untreated.....	Untreated...	1,770....	0	0
181.....	550.....	1,125....	100	99
182.....	650.....	1,060....	100	99
185.....	750.....	775.....	100	99
187.....	850.....	155.....	100	99
189.....	950.....	240.....	99	98
190.....	1,130....	170.....	100	98
195.....	1,260....	260.....	100	98
199.....	1,430....	360.....	100	99
204.....	1,740....	1,165....	99	97
210.....	2,100....	960.....	98	97
215.....	2,400....	Burn through		

Example VI

To permit the study of the effect of higher levels of discharge current on the treatment, a series of film samples made from the same film forming grade of polyethylene as heretofore referred to (DYNH-3) and of the same lot as used in Example I above, were treated by an apparatus in which the grounded roll was covered with 10 layers of 1 mil thick Mylar (polyethylene terephthalate) to prevent burn-through despite the higher current levels. The treatments were carried out substantially immediately following formation of the film samples by an extrusion apparatus. The results were as follows:

Primary Voltage in Volts	Primary current in milliamps	Heat Seal Strength (Gms.)	Ink Adhesion, Percent Retention	
			Dry	Wet
Untreated.....	Untreated...	about 1,200..	0-5	0-5
155.....	520.....	620.....	100	100
160.....	850.....	1,098....	100	100
200.....	1,210....	930.....	100	100
225.....	1,500....	640.....	100	40
258.....	1,940....	540.....	100	20

A "burn-through" value for this sample of film on an unwrapped ground roll was 1200 milliamps.

A comparison of the results set forth in the immediately preceding table with the results set forth in the table under Example I will show that the distinct increase in the heat seal strength was achieved at all current levels with the grounded roll wrapped, though at current levels above that at which burn-through occurred when the non-wrapped grounded roll was employed no increase in heat sealability was effected with the wrapped grounded roll; that is, it would appear that no advantage would be gained, even when employing a wrapped grounded roll that would prevent burn-through by employing current levels in the electrode circuit above that at which burn-through would occur were no wrapping of the grounded roll employed. It should be noted also from the table of this example that the percentage retention of ink, when subjected to the "Wet" ink adhesion test decreased considerably when the current level of the treatment was increased (grounded roll wrapped) beyond the value at which burn-through would occur were the grounded roll unwrapped.

From the above, it will be clear that the maximum heat sealability characteristic is obtained when the current level of the electrode circuit in an electric glow discharge treatment of polyethylene (when employing either a wrapped or unwrapped grounded roll) is just below

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the current level at which a burning through of the polyethylene would occur with an unwrapped grounded roll. In addition, it will be noted that if the current level is increased from a relatively low value toward the burn-through value, the heat sealability achieved passes through a minimum increasing upon further increases of the current level in the electrode circuit to a treated film maximum at the burn through level.

While, as indicated, the heat sealability characteristic obtained by the use of a wrapped grounded roll at substantially all current levels is substantially greater than that obtained at the same current levels and under the same conditions with an unwrapped grounded roll, we have to date found no wrapping material, including Mylar, which will be satisfactory for a greater period of continuous use than about 24 hours. Hence our recommended treatment still employs an unwrapped grounded roll.

As a result of the above, our recommended procedure for the treatment of polyethylene film or the like, to increase the anchorage or adherence characteristics thereof without at the same time destroying the heat sealability characteristic thereof, and in fact to obtain the maximum heat sealability characteristics thereof (in treated condition, of course) is to raise the current level in the electrode circuit to the point at which burn-through occurs and then to reduce such current level not more than about 15% but at least enough to assure that burn-through will not occur despite the normal variations in film thickness and character encountered in the normal commercial operation. Generally a reduction of about 5% will give such assurance. If a wrapped grounded roll is being employed, it would be preferable to use an auxiliary device which has been previously correlated with the main apparatus including the wrapped grounded roll, for determining the current level to be employed in the main apparatus. In either case, of course, that is, both when using a wrapped grounded roll and when using an unwrapped grounded roll, the current level setting procedure should be carried out with each different lot of polyethylene to be treated.

The phenomena leading to the advantages of the method of this invention are believed to be based on the existence of a number of different simultaneous effects of the electric discharge treatment on the polyethylene, including oxidation, depolymerization, cross-linking of different molecular chains, and mechanical erosion of the surface. It is believed that at least some of these effects were responsible for impaired heat seal properties and it is postulated that at the higher treating currents just below the burn-through value, such effects are minimized while the effects which do not impair the heat seal properties are favored.

We claim:

The method of electrically treating a surface of a polyethylene structure to improve its adhesion to printing inks with optimal retention of the heat-sealability characteristics thereof which consists of establishing an alternating current electrical glow discharge between a pair of spaced electrodes, passing a polyethylene structure between said electrodes to subject a surface thereof directly to said discharge, and maintaining the electrical current in said discharge at a value between substantially 5% to 15% below the lowest value at which said discharge is accompanied by electrical arcing through said structure.

References Cited in the file of this patent

FOREIGN PATENTS

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