Jan. 26, 1954 G. J. GOEPFERT ET AL 2,667,436

PRESSURE SENSITIVE ADHESIVE COATED SHEET MATERIAL

Filed Sept. 21, 1950

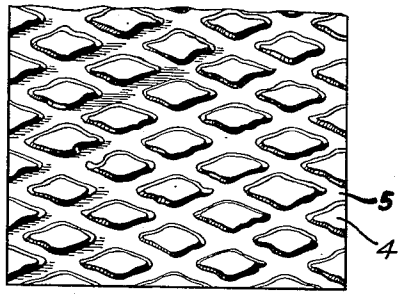


FIG. 1

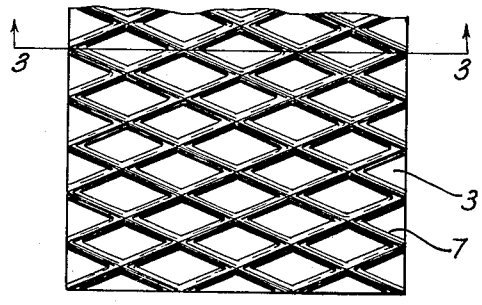


FIG. 2

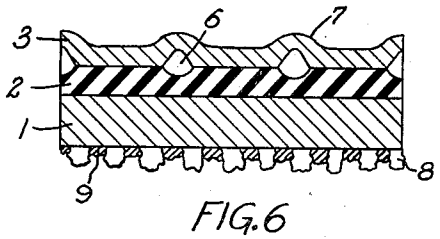


FIG. 6

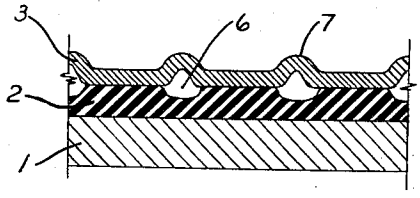


FIG. 3

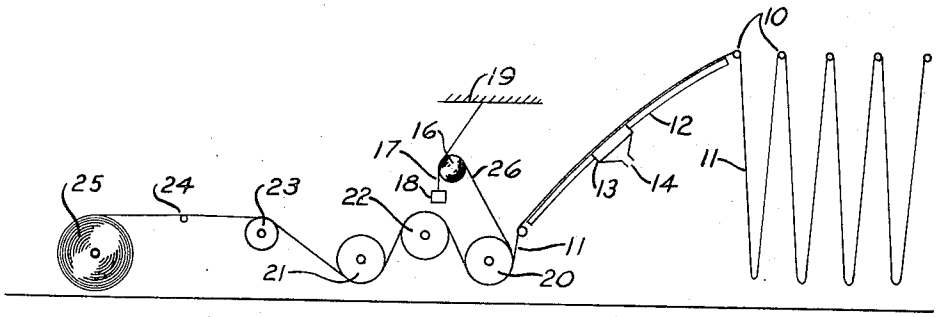


FIG. 4

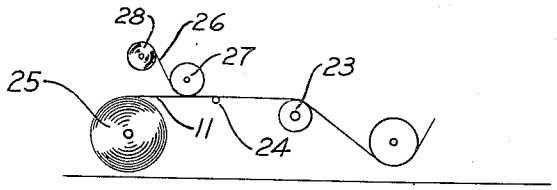


FIG. 5

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PRESSURE SENSITIVE ADHESIVE COATED SHEET MATERIAL

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9 Claims. (Cl. 154—53.5)

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This invention relates to sheet material coated on at least one side with a pressure sensitive adhesive and to a method of making the same. More specifically, it is directed to the production of such material wherein at least a part of the adhesive is provided in the form of a pattern consisting of raised nodules separated by continuous depressed channels formed by applying to the adhesive a sheet of embossed polyethylene.

Products with which this invention are concerned are made in numerous forms and used for many purposes. In some instances the material is wound into rolls in which case it is customary to treat the back of the sheet material to prevent the pressure sensitive adhesive from sticking to it. In other cases where the sheet material is coated on both sides or where it is marketed in special forms such as sheets coated on one side with a non-slip material it is customary to apply a protective sheet to the pressure sensitive adhesive. One very commonly used form of protective sheeting is Holland cloth, though other such materials have been proposed and used such as a combination of thin paper and print cloth or paper coated with shellac. Where such sheets are employed it is difficult to apply them to the adhesive surface in such a manner that they are free from wrinkles. Furthermore, these materials are opaque and make it impossible to inspect the pressure sensitive adhesive coating after the protective sheet has been applied. Difficulties are also experienced from excessive sticking of the protective sheet to the adhesive and many of these sheets which have been used are not sufficiently impervious to air with the result that the adhesive is improperly protected against oxidation. If oxidation occurs the adhesive loses its tack and becomes stiff and unsatisfactory.

We have discovered that pressure sensitive adhesive can be very satisfactorily protected by the use of a pigmented film of polyethylene which has been embossed to provide parallelograms which lie in substantially the same plane and which are separated from but attached to each other by raised ribs which are relatively narrow and shallow. We prefer to emboss with rhombs which are so formed that the ribs between the rhombs form continuous lines extending to the sides of the protective sheet.

We have further discovered that it is important that the protective sheet be applied with the rhombs next to the pressure sensitive adhesive film and the ribs extending outwardly. If the sheets are applied with the ribs downward in contact with the pressure sensitive adhesive, gas pockets form when the sheet is applied and these pockets develop into blisters or bubbles which make the product unsatisfactory from a commercial standpoint. On the other hand, if the sheet is applied with the ribs outward and if there is the proper combination of tackiness of the pressure sensitive adhesive, thickness of the polyethylene film, depth of embossing of the polyethylene film, plasticity of the pressure sensitive adhesive, and pressure applied when the protective film is placed on the pressure sensitive adhesive surface, the pressure sensitive adhesive becomes attached to the protective sheet only at the parallelograms or rhombs and in addition the pressure sensitive adhesive film takes on a more or less perfect mirror image of the protective sheet. In such case a substantial portion of the pressure sensitive adhesive surface consists of raised portions which are adhesively attached to the parallelograms on the protective sheet, spaced from each other by depressed portions which register with the ribs on the protective sheet and provide channels of substantial cross-section through which any entrapped gases can escape.

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Depending upon the conditions, the pattern on the pressure sensitive adhesive may be practically a reproduction in reverse of the pattern of the protective sheet. If conditions are not ideal, the reproduction of the parallelograms becomes imperfect, first being such that the edges of the raised rhombs of the pressure sensitive adhesive approach each other and the channels in the adhesive become correspondingly reduced in area. In the extreme case the pressure sensitive adhesive will be shaped to a reproduction rather than a mirror image of the protective sheet. In such case the pressure sensitive adhesive consists of depressed parallelograms and raised ribs. In other words, in this extreme case the protective sheet contacts the pressure sensitive adhesive over substantially its entire area. Such condition is undesirable because it blocks up the escape of the gases and tends to result in blisters such as are obtained when the embossed film is applied with the raised ribs in contact with the pressure sensitive adhesive.

We have further discovered that a pressure sensitive adhesive film can be produced which is in the form of raised rhombs separated by connecting channels on sheet material coated in long webs by applying the embossed protective sheet to the pressure sensitive adhesive as described, removing the protective sheet, and winding the

material into rolls. Such product has the advantages that there is a substantial reduction in the tendency for the pressure sensitive adhesive to stick to the back of the sheet material and that such product can be caused to adhere to other materials without wrinkling or forming blisters. If a sheet is coated with pressure sensitive adhesive wherein the adhesive is in the form of a substantially smooth film there is a tendency for the sheet to buckle or even to form blisters when it is attached to some other object. With the improved design of pressure sensitive adhesive obtainable according to this form of our invention such tendency is substantially reduced and it is very easy to spread out the sheet material so that it will be perfectly smooth. If material is applied under considerable pressure the sheet adheres firmly whereas if it is desired to have the sheet attached temporarily this can be done by applying light pressure such that only the rhombs contact the other object and the sheet can be much more easily detached.

We have illustrated our invention by reference to drawings wherein:

Fig. 1 is a plan view of a portion of the pressure sensitive adhesive as it is formed according to the preferred embodiment of the invention;

Fig. 2 is a plan of a piece of sheet material covered with the protective film;

Fig. 3 is a section along the lines 3—3 of Fig. 2;

Fig. 4 is a diagrammatic side elevation of a portion of a form of apparatus suitable for carrying out one modification of our invention;

Fig. 5 is a diagrammatic side elevation of a portion of a form of apparatus suitable for carrying out another modification of our invention; and

Fig. 6 is a section of the product made according to another embodiment of the invention.

Referring particularly to Fig. 3, the product consists of a backing 1 which may be paper, cloth, plastic or other suitable sheet material coated on one side with a pressure sensitive adhesive film 2 which is covered by the protective sheet of polyethylene 3. As is illustrated in Fig. 1, the pressure sensitive adhesive consists of raised rhombs 4 separated by channels 5. When the protective sheet is on the film, as shown in Figs. 2 and 3, the channels 5 register with the openings 6 below the ribs 7 of the protective sheet.

The embodiment shown in Fig. 6 is similar to that shown in Fig. 3 except that the face of the sheet material opposite the one which is coated with the pressure sensitive has a layer of granular material 8 attached to it by an adhesive 9.

In Fig. 4 we have illustrated a form of apparatus suitable for carrying out the process of one modification of our invention. Referring thereto, reading from left to right, the apparatus consists of sticks 10 in a conventional festooning apparatus on which the coated material 11 is suspended and from which it travels over a toboggan 12 provided with a suction box 13 connected through opening 14 to a source of suction not shown in the drawing. The protective sheet material is provided in the form of a roll 16 over which is hung a drag 17, such as a sheet of canvas or other suitable cloth, to the bottom of which is attached a weight 18. The upper end of the drag is adjustably attached to framework 19 so that the area of the drag which contacts the sheet material, and correspondingly the amount of the drag, can be varied. The apparatus also contains tension rolls 20 and 21 between which there is an idler roll 22, counterroll 23, idler roll 24 and wind-up roll 25.

In employment of the apparatus the adhesive coated sheet 11 is brought down over toboggan 12 and into contact with the polyethylene film 26. The combination is then threaded around rolls 20, 22, 21, 23 and 24 and fastened to wind-up roll 25. Suction is applied to the suction box 13 to assist in effecting tension on the sheet material and tension is also controlled by the tightness of a belt, not shown in the drawings, which drives tension rolls 20 and 21 from the same source of power as is applied to the wind-up roll 25, also not shown in the drawings. The wind-up roll, the tension, idler and counterrolls, toboggan and festoons are all conventional in the coating industry and the manner in which this form of apparatus is used is well known in that art.

The apparatus illustrated in Fig. 5 is adapted for the manufacture of sheet material coated with pressure sensitive adhesive wherein the adhesive is in the form of raised rhombs as illustrated in Fig. 1 but no protective sheet is used. That apparatus is similar to the apparatus of Fig. 4 up to the idler roll 24 but differs from the apparatus of Fig. 4 by the provision of an idler roll 27 and an additional wind-up roll 28. In making this form of product the sheet material is coated and covered with the protective sheet as in making the product according to the process of Fig. 4 but when the combined sheet reaches roll 27 the protective sheet is separated from the adhesive coated sheet and pulled up around idler roll 27 and attached to wind-up roll 28.

As stated, we have found that it is necessary to control numerous factors in order to obtain this ideal condition of the pressure sensitive adhesive film. In general we have found that it is desirable to have the pressure sensitive adhesive relatively plastic when the protective sheet is applied. This may be done by having the sheet material warm when the protective sheet is applied as by assembling the protective sheet on the pressure sensitive adhesive film when the product comes from the oven in which the sheet material is heated to remove solvents from the pressure sensitive adhesive coating.

We have also found it necessary to maintain at least a minimum ratio between the depth of embossing of the protective sheet and the thickness of the pressure sensitive adhesive film although this minimum is in turn somewhat dependent upon the pressure applied to the protective sheet when it is placed over the pressure sensitive adhesive film. Furthermore, there is a somewhat critical ratio between the thickness of the polyethylene film and the depth of the embossing.

The thickness of the pressure sensitive adhesive film is usually determined by limitations of the use for which the product is made as well as the kind of sheet material employed. For example, in making a sheet material for non-slip purposes where it is customary to coat the cloth backing on one side with granular material such as abrasive grains to make a product somewhat similar to abrasive cloth and on the other side with pressure sensitive adhesive, it is desirable to employ adhesive films of the order of .005 inch to .008 inch in thickness. On the other hand, where sheets are being made as by coating a sheet of paper with the pressure sensitive adhesive to provide products to be used as labels the pressure sensitive adhesive film may be thin-

ner. In general we have found that it is necessary to employ adhesive films at least about .003 inch thick in order to obtain satisfactory adhesion although this thickness is determined somewhat by the character of the adhesive used.

Where the pressure sensitive adhesive film is relatively thick the embossing should be correspondingly deep since we have found that a lightly embossed sheet when applied to a thick film is likely to reproduce itself in the film rather than form the desirable mirror image. While we do not wish to be bound by any theories it has been suggested that this mirror image results when conditions are such that the ribs on the protective sheet are pressed against the pressure sensitive adhesive film so lightly that the resilience of the polyethylene causes the ribs first to become embedded in the pressure sensitive adhesive when the sheet is pressed against the film and then to retract when the pressure is released thereby forming these raised parallelograms in the pressure sensitive adhesive which are spaced between depressed lines matching with the ribs on the protective sheet. In any event, by proper selection of the combination of the characteristics and structure described above along with suitable conditions of temperature and pressure the desirable configuration of the pressure sensitive adhesive film is obtained.

We will now illustrate our invention by specific examples. It is to be understood that the examples are not limitative but are merely descriptive of products and combinations which will result in the product of our invention.

Example 1

A drill cloth of such weight that a piece 32 inches wide contains 2.58 yards per pound, having a count of 72 x 48 and 14's warp was filled with 8 pounds per sandpapermaker's ream of 480 sheets 9 x 11 inches of a phenol-formaldehyde resin. The cloth was then coated with 28 pounds per ream of 60 grit silicon carbide attached by a phenol-formaldehyde resin. Methods of making such products are well known in the abrasive industry and will not be further described since they form no part of this invention. One suitable method, for example, is that described and claimed in U. S. Patent No. 2,184,896.

The back of the cloth was then coated with 6 pounds per ream of a priming adhesive which was heated for 1 hour at 175° F. to remove the solvent and a coating of pressure sensitive adhesive to the extent of 30 pounds wet weight was applied over the priming coat. This was then heated for 2 hours at 175° F. to remove the solvents.

The priming adhesive contained approximately 50% solids suspended in water, the solids being of the following composition:

	Per cent
Natural rubber latex	60
Clay	20
Antioxidant	5
Methyl cellulose	5
"Hercolyn"	10

The pressure sensitive adhesive was a solution containing 30% solids dissolved in gasoline. The solids composition was as follows:

	Per cent
"GRS" rubber	45
"Staybelite" resin	20
"Hercolyn"	8
Zinc oxide	27

"GRS" rubber is a copolymer of butadiene with styrene manufactured according to a Government specification. The "Staybelite" resin is hydrogenated rosin and the "Hercolyn" is hydrogenated methyl abietate. "Staybelite" and "Hercolyn" are manufactured by Hercules Powder Company.

The pressure sensitive adhesive solution was prepared by milling the solid materials described to obtain a homogeneous mixture and then dissolving this mixture in the gasoline. 35 pounds per ream wet weight were applied which when dried left a film .007 inch thick.

After the solvent had been removed from the pressure sensitive adhesive the product was taken from the drying racks where it was heated in festoon form and was covered with a protective film of pigmented polyethylene which had a molecular weight in the range 15,000 to 25,000 and which had first been formed into a sheet .0055 inch thick and then embossed in the form of rhombs the sides of which were approximately $\frac{1}{16}$ inch long. The rhombs were formed with the long diagonal lengthwise of the sheet and of such shape that the greater diagonal was twice that of the lesser. The sheet had 11½ rhombs to the inch crosswise of the sheet and 5 to the inch lengthwise. The depth of the embossing was approximately .002 inch. Its molecular weight was approximately 20,000 and it contained approximately .2% of a blue dye consisting of a copper salt of a phthalocyanine dye and about 1% of zinc oxide to increase the opacity of the film.

The protective sheet was applied in the apparatus illustrated diagrammatically in Fig. 4. Sufficient tension was applied to the roll of embossed polyethylene to cause it to feed evenly but without substantial tension onto the pressure sensitive adhesive surface. The tension in the adhesive coated sheet material was first adjusted so that the combined material traveled through the rolls with a slight inclination to wander from side to side and was then increased just slightly to prevent this sidewise motion and result in a straight continuous flow around the rolls 20, 22, 21, 23 and 24 and up to the final wind-up roll 25.

The tensile strength and stretch of the polyethylene film were determined both lengthwise and crosswise on a Scott paper tester in which the jaw was pulled at the rate of 12 inches per minute. Tests were made on strips 1 inch wide with the jaws 1 inch apart at the start. Under these conditions of test the material tested lengthwise stretched 8 inches and broke under a load of 4 pounds. The cross stretch was 6 inches and the cross strength was 3½ pounds.

Example 2

130 lb. rope cylinder paper of a type commonly employed in the abrasive industry was first coated with a priming coat as described in Example 1, then with the pressure sensitive adhesive of Example 1, the wet weight of the pressure sensitive adhesive coat being 20 pounds per ream. When dry this left a film approximately .004 inch thick.

The protective sheet employed was .004 inch thick and had been embossed in the form of rhombs the sides of which were $\frac{1}{16}$ inch long. The rhombs were formed with their long diagonals lengthwise of the sheet and of such proportions that the long diagonals were approximately twice that of the lesser. The sheet had 14 rhombs to

the inch crosswise and 6½ to the inch lengthwise. The depth of the embossing was .0015 inch. A strip tested as described in Example 1 pulled lengthwise broke under a load of 4 pounds and stretched 4 inches. Crosswise it broke at 3 pounds and stretched 3 inches. It had a molecular weight of approximately 20,000 and contained approximately .2% of a green dye consisting of a copper salt of a phthalocyanine dye and about 1% of zinc oxide to increase the opacity of the film.

The sheet was applied as described in Example 1 but was removed in the apparatus illustrated in Fig. 5 by stripping it from the pressure sensitive adhesive film around idler roll **27** and wound up on roll **28**. The product obtained had the pressure sensitive adhesive in substantially the shape illustrated in Fig. 1. The protective sheet was easily stripped from the pressure sensitive adhesive and the product was readily unwound from the roll form in which it was prepared.

We have stated that there are certain critical conditions required in order to produce the product of the raised rhombs. Because of the differences between various compositions of pressure sensitive adhesive it is impractical to attempt to specify the exact conditions for all the various modifications in which the invention may be employed. We have given two specific examples and have there described conditions which resulted in the desired configuration of the pressure sensitive adhesive film. Following are statements of the conditions which tend to promote formation of this desired pattern.

1. With increased thickness of pressure sensitive adhesive it is necessary to increase the depth of embossing of the protective sheet.

2. For any given plasticity of the pressure sensitive adhesive, which increases with the temperature, and for any given protective sheet elasticity, there is a maximum pressure at which the sheet can be applied to the film without causing a reproduction rather than a mirror image. The pressure should preferably be only sufficient to bring out the mirror image as increases in pressure cause first a tendency for closure of the channels and eventually a reproduction.

3. The embossed film must be applied with the ribs out.

4. The depth of embossing should be about 25–50% of the thickness of the protective sheet.

5. In general the thickness of the protective sheet should not be less than the thickness of the pressure sensitive adhesive film nor more than about .007 inch for pressure sensitive adhesive films of the thickness usually employed.

6. While the desired pattern can be reproduced by the use of protective sheets embossed with parallelograms, we have found that where the parallelograms are squares there is some tendency for the formation of blisters, possibly because one-half of the channels run lengthwise of the sheet whereas when the embossing is in the form of rhombs the channels all extend to the edges. For this reason we prefer to use rhombs and in general have found that it is desirable to have the rhombs with the long diagonals running lengthwise of the film and being approximately twice as long as the short diagonals.

In the specific examples we have described the use of zinc oxide to increase opacity and a particular green pigment which is sold by the Harmon Color Works of Patterson, Pennsylvania, under the trade-mark "Opaline G1." Pigments for this purpose are well known and are manufactured by numerous concerns such as the Du Pont Company and the Imperial Paper and Color Corp. of Glens Falls, New York. One most common type of pigment is the salt of a phthalocyanine which can be obtained in either a blue or a green color, typical pigments being Du Pont's "Monastral Green" or "Ramapo Green," or Harmon's "Luna Blue" and Imperial's "Red F 1869" and "Sherwood A 4436." Other pigments may be used, including both organic and inorganic materials. Where organic pigments are used we employ them in the range of about .05 to 2% and where inorganic pigments are used we may use from about 1% to about 10%. In the examples we have described the use of 1% of zinc oxide to increase the opacity of polyethylene film. The zinc oxide may be omitted or, if some increase in opacity above that imparted by an organic pigment is desired, inorganic pigments either alone or along with organic pigments may also be used.

We have illustrated our invention by reference to two specific examples and have set forth certain principles to be applied in carrying out the invention. We have also set forth a suggested explanation of why this unexpected pattern is formed. It is to be understood that our invention is not restricted to the specific examples and that we are not to be bound by the theories as to why the process results in the product but, rather, that the invention is to be of the scope set forth in the appended claims.

We claim:

1. As a new article of manufacture, sheet material having a film of pressure sensitive adhesive adhesively attached to at least one side, and a protective sheet consisting essentially of pigmented polyethylene embossed to form parallelograms which lie substantially in the same plane and which are separated from but attached to each other by raised ribs adhesively attached to the outer surface of the pressure sensitive adhesive film with the ribs on the outer side, the pressure sensitive adhesive film being in large part in the form of parallelograms which are separated by channels in which the pressure sensitive adhesive coating is thinner than that of the parallelograms and are attached to the parallelograms on the protective sheet, the channels in the pressure sensitive adhesive and the ribs on the protective sheet forming channels which communicate to the edge of the sheet through which gases which might be entrapped between the pressure sensitive adhesive film and the protective sheet can escape whereby the protective sheet is substantially free from blisters and bubbles.

2. Article as claimed in claim 1 wherein the pressure sensitive adhesive film is about .003 to .008 inch thick and the polyethylene sheet is about .003 to .007 inch thick.

3. Article as claimed in claim 2 wherein the depth of the embossing of the protective sheet is about 25–50% of the thickness of the said sheet.

4. Article as claimed in claim 3 wherein the parallelograms are rhombs.

5. Article as claimed in claim 4 wherein the thickness of the pressure sensitive adhesive film is about .004 to .007 inch, the thickness of the protective sheet is about .0045 to .0055 inch, the length of each side of the rhombs is approximately $\frac{1}{16}$ inch and the depth of embossing is about .0015 to .002 inch.

6. Article as claimed in claim 5 wherein only

one side of the sheet material is coated with pressure sensitive adhesive and the other side is provided with a coating of granular material.

7. Article as claimed in claim 6 wherein the polyethylene has a molecular weight of about 15,000 to 25,000.

8. Article as claimed in claim 1 wherein only one side of the sheet material is coated with pressure sensitive adhesive and the other side is provided with a coating of granular material.

9. As a new article of manufacture, sheet material comprising a backing having a substantially planar surface and a coating of pressure sensitive adhesive attached to said surface, said coating consisting of a multiplicity of parallelograms separated from each other at their outer surfaces but joined at the surface of the backing by thinner films of the pressure sensitive adhesive, the thinner films being in the form of substantially continuous channels which provide passageways for gases whereby when the pressure sensitive adhesive coated surface is pressed onto another surface to cause attachment thereto the air which might be entrapped between the two surfaces escapes through the channels and the sheet material is attached to the other surface smoothly and substantially free from wrinkles.

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