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C. W. STILLWELL
GUMMED SHEET MATERIAL

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Fig. 1

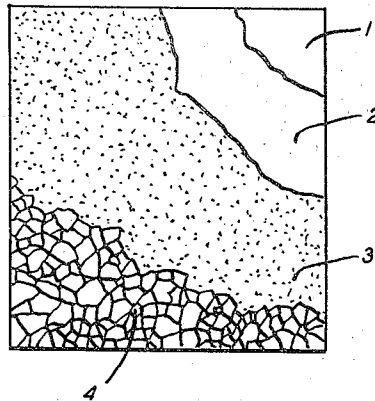
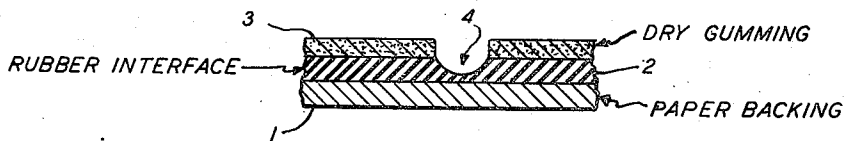


Fig. 2



Inventor:
Charles W. Stillwell
By Roberts, Gushman & Woodbury
his Attys.

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GUMMED SHEET MATERIAL

Charles W. Stillwell, Framingham, Mass., assignor to Dennison Manufacturing Company, Framingham, Mass., a corporation of Massachusetts

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11 Claims. (Cl. 117—41)

This case is a continuation-in-part of Serial No. 255,448, filed February 9, 1939.

This invention relates to gummed paper and other sheet material coated with a film of adhesive which is normally non-tacky but which quickly becomes tacky when remolstened. Ordinarily the film comprises dextrin, animal glue, fish glue or the like, applied in fluid form by a suitable coating process and then dried. Due to shrinkage of the coating during the drying process the paper tends to curl toward the coated side. To counteract this tendency it is customary to draw the paper under tension over an edge to break the gumming into multitudinous minute areas separated by fine cracks, this process being called mechanical breaking in contradistinction to self-breaking produced by holding the paper taut while the coating dries. This mechanical breaking not only constitutes an additional operation in the process of manufacture but, owing to the necessary tension on the paper, it often tears, resulting in loss of time and material. Moreover, the broken paper still tends to curl to some extent if subjected to substantial change in humidity.

The principal objects of the present invention are to provide gummed paper which is substantially non-curling, which remains in this condition throughout varying conditions of atmospheric temperature and humidity, and which need not be broken mechanically.

According to this invention the gum coating is not applied directly to the paper but is applied over a preliminary coating characterized by sufficient give to prevent curling after the paper is broken and to permit satisfactory self-breaking by holding the paper taut while drying. The preliminary coating is hereinafter referred to as the interface.

If ordinary gummed paper is held taut while drying the gum coating breaks itself by shrinkage but the coating breaks up into such a large number of extremely minute areas that the cracks therebetween are correspondingly numerous and therefore almost imperceptibly fine. Indeed the cracks are so fine they are self-healing with changes in humidity, that is they close up and the minute areas fuse together again, thereby fully restoring the tendency to curl. As I have now discovered, this unsatisfactory behavior of ordinary gummed paper is due to the fact that the coating is anchored directly and firmly to the paper so that substantially no relative movement between coating and paper can take place edgewise of the paper.

I have also discovered that a yielding interface will permit self-breaking into areas which, while still minute, are large enough to produce cracks too wide to be self-healing. Indeed the size of the areas can be regulated throughout wide limits by controlling the degree of give of the interface. Were the gumming floated on an interface of liquid having unlimited give, the gumming could shrink without cracking and without curling the paper. Passing from this hypothetical extreme into the practical realm of interfaces having less give and a practical degree of strength, the interface may be compounded to afford any desired size of areas down to the point where the cracks are self-healing. For most purposes the cracks should be as fine as possible without being self-healing, the minute areas of gumming being correspondingly small, particularly if the paper is to be printed. Wide cracks mitigate against good printing even though the cracks are on the unprinted side.

The principle upon which the yielding interface attains the aforesaid objects I believe to be as follows. Considering the shrinkage of the gum coating while drying as setting up stresses similar to those which would result from a corresponding expansion of the paper, these forces tend to produce relative slippage between paper and gumming when the paper is held taut during the drying operation. As slippage takes place the interface is distorted, creating therein strains whose magnitude depend upon various factors including the elasticity and viscosity of the interface. When the strains build up to the breaking point of the film of gumming, the film begins to crack up, first into large islands and then, as shrinkage of the gumming continues, the large islands progressively break up into smaller islands until the film is completely dry. When the film breaks up the stretched gumming of each individual island contracts, thereby widening the cracks between and removing some of the strains in the film of gumming. As the islands continue to dry, further shrinkage and cracking occur until the gumming is substantially dry. With much give in the interface, much shrinkage can take place before the gumming breaks up, resulting in large islands and wide cracks. With little give in the interface, little slippage can take place and the film ultimately breaks up into small islands.

While the interface may comprise a plastic composition it is preferably formed of rubber-like material, that is rubber or a material such as plasticized polyvinyl chloride having mechani-

cal properties similar to rubber, characterized in that, while it may be deformed through small distances by small forces, its resistance to deformation builds up rapidly with increased deformation. Such an interface yields easily through distances which, though small, are large enough to permit the paper to remain flat after the gum film has cracked up as aforesaid. At the same time the interface is strong enough to withstand the practical uses to which the paper is normally subjected. A rubber-like interface is also usually preferable to a plastic interface because, other factors being equal, it causes the gum film to break up into a smaller pattern.

While the interface preferably comprises natural rubber, such as pale crepe rubber, it may also comprise other plastomeric and/or elastomeric compositions including rubbers such as balata, gutta-percha, gutta-siak, pontianak, guayule, synthetic elastomers (see H. L. Fisher, I. & E. Chem. 31, 941, 1939) such as Vistanex (polyisobutene), Buna (a copolymer of butadiene with styrene or acrylonitrile), Neoprene (polymerized chloroprene), Thiokol (polyethylene polysulphide), and any one of the following compositions when plasticized to the proper degree of deformability hereinafter defined: polystyrene, polyvinyl chloride, polyethylidene chloride, polyvinyl alcohol, polyvinyl esters such as polyvinyl acetate, copolymers of vinyl chloride and vinyl esters such as the copolymer of vinyl chloride and vinyl acetate, polyvinyl formal, actetal, butyral, nitrocellulose, cellulose acetate, chlorinated rubber, polyacrylates or methacrylates, such as polymethyl, ethyl, propyl, butyl, acrylate or methacrylate, ethyl cellulose, starch acetate, shellac, elastic sulfur, casein, soya bean protein, zein, gelatine, glue or super polyamides.

The interface coating may be applied by any of the well-known methods, either from a solution or in molten condition, but it is preferably applied with a solvent such as benzol which does not cause the paper to swell. After the interface coating has been applied the gum coating of dextrin, animal glue, fish glue, or the like may be applied in any of the well-known ways, as by a doctor blade or a roll.

The following are a few typical examples of compositions suitable for use in forming the interface coating, the numbers indicating the proportional parts by weight:

(1)	
Pale crepe rubber.....	100
White factice.....	50
Benzol	300

(2)	
Pale crepe rubber.....	100
White factice.....	50
Hydrogenated rosin.....	25
Benzol.....	300

(3)	
Water white rosin.....	100
Dibutyl phthalate.....	20
1000 second nitrocotton.....	10
Ethyl acetate.....	200

(4)	
Maizite (the prolamine, zein).....	100
Bleached de-waxed shellac.....	100
Dibutyl tartrate.....	75
Ethyl alcohol.....	250

(5)	
Vistanex #6 (a polymer of isobuten).....	100
Benzol.....	200

(6)	
Washed and dried <i>Coquilariana balata</i>	100
Hydrogenated rosin.....	50
Gutta-percha resin.....	25
Pale crepe rubber.....	5
Benzol.....	400

(7)	
Pontianak.....	100
Aroclor #1262 (a polymerized chloro bi-phenyl).....	25
Crepe rubber.....	10
Benzol.....	150

(8)	
Pale rubber crepe.....	100
Benzol.....	900

(9)	
Ethyl cellulose (20 cp. standard ethoxy).....	90
Dibutyl phthalate.....	10
Toluol.....	280
Ethanol.....	120

(10)	
Ethyl cellulose (40 cp. standard ethoxy).....	15
Di (O-xenyl) monophenyl phosphate (Dow plasticizer No. 6).....	85
Toluol.....	20

In the case of the first two examples the solid components are milled together while hot for about twenty minutes before being dissolved in the benzol.

As indicated by the aforesaid illustrative examples, a wide variety of materials may be used to produce an interface of suitable plastic and/or elastic quality to achieve the desired results. They vary over a considerable range in the degree of plasticity or elasticity, and have various effects upon the relative flatness of the paper. However, all of these materials have the property of being deformable when subjected to forces of the order of magnitude of those developed by the contraction of a drying glue film spread upon them. Thus they provide a buffer layer between the gumming and the paper such that the strain resulting in unequal expansion and contraction of the surface gumming and the paper, when subjected to changing conditions of humidity, may be readily dissipated in the comparatively local areas into which the surface gum has broken itself.

For gummed sheets which are self-breaking and substantially non-curling (flat enough for ordinary usage) the interface should be at least as deformable as a film having an elongation of 4% within 20 seconds at a load of 350 kg. per sq. cm. (determined by taking the stress-strain curve of a cast film by the method of Bass and Kauppi in Industrial and Engineering Chemistry, vol. 29, No. 6, June 1937, p. 678 et seq.); any interface having substantially less deformability does not produce sufficient breaking of the gumming to cause the sheets to lie flat. Aforesaid No. 9 is a suitable composition for making an interface having this lower limit of deformability. On the other hand the interface should be no more easily deformable than a composition having at 25° C. a penetration of 300 sec. (taken on a cone-type penetrometer as described in H. A. Gardner's A Physical and Chemical Examination of Paints,

Varnishes, Lacquers and Colors, 7th ed., Washington, D. C., Institute of Paint and Varnish Research); substantially more deformable compositions are too weak to provide an adequate bond between gumming and backing for most purposes. Aforesaid No. 10 is a suitable composition for making an interface having this upper limit of deformability. For average uses best results are obtained with an interface having an elongation of approximately 1000% in 20 sec. at a load of 100 kg. per sq. cm. A suitable composition for making such an interface is aforesaid No. 8.

The upper and lower limits of interface deformability must be determined by different methods for the reason that any method applicable to the harder compositions at the lower end of the permissible range is inapplicable to the softer compositions at the upper end of the range, and vice versa. While the aforesaid elongation method is the one favored by the majority of experts in the art for determination of the deformability of the harder compositions, it is inapplicable to the compositions at the other end of the range because they are too soft and tacky; and while the penetration method is the one best suited to the softer compositions it is unsuited to the compositions at the other end of the range between the penetration is too slight.

While the amount of interface may vary from one pound or less per ream (500 sheets 20" x 25") upwards, for most purposes it is preferably in the region of two to three pounds per ream, and seldom if ever does it need to be more than approximately six pounds per ream. The amount of gumming may vary from about two pounds per ream up to twelve pounds or even more. Any remoistenable gumming can be used; for example, a low-viscosity comparatively low strength glue such as is normally used in gummed paper manufacture, and which may have a gel strength of about 50 grams in 12.5% solution taken on a Bloom gelometer, according to Standard Methods (revised) for Determination of Viscosity and Gel Strength of Glues, Industrial and Engineering Chemistry, analytical edition 2, 348 (1930), or a high strength gelatine glue such as is used in woodworking and which may have a gel strength of 500 grams or more, taken according to the same technique. If the gumming is too strong or too thick for the underlying interface it is not self-breaking or merely breaks into a few islands which are too large to prevent curling; the interface should be strong enough in relation to the strength of the gumming to produce self-breaking of the gumming. The break pattern (i. e., the size of the islands and the width of the cracks) increases with increase of any one or more of the following factors: Interface thickness, interface deformability, gumming thickness and gumming strength (up to the point where the gumming is too strong to break). It follows that any one of these factors may be increased without increasing the size of the break pattern by correspondingly decreasing any one or more of the other factors. For example the strength of the gumming may be reduced by adding a plasticizer, such as glycerine, sorbitol, "Sorbitan" (a technical grade of mono-anhydro sorbitol), triethanol amine or a finely dispersed filler, such as latex, resin emulsions such as an alkyd resin emulsion, mica, clay, etc., or both. In general weak and/or thin gummings can be used with interfaces which are thin and/or near the lower end of the range of deformabil-

ity; and conversely interfaces which are thick and/or very deformable permit the use of thicker and stronger gummings. For example, when using the aforesaid interface No. 8 with the aforesaid weak and strong gummings respectively, the following thicknesses, in terms of pounds per ream, are recommended:

	Weak gumming			Strong gumming	
Gumming.....	2-3	5-6	10-12	2-3	5-6
Interface.....	0-7	2-3	2-3	2-2.5	3-4

For the purpose of illustration a typical product of the present invention is shown in the accompanying drawing in which

Fig. 1 is a face view of a piece of gummed paper showing the gum film broken in sections or islands of the preferred size; and

Fig. 2 is an enlarged section showing how the interface stretches at the cracks in the gumming.

As shown in the drawing the product comprises a paper backing 1, a rubber interface 2 and sections 3 of dry gumming separated by cracks 4.

The following is an outline of what I believe takes place as the glue film dries. The drying of the glue results in contraction of the film. The easily deformed interface permits this at first but, with rapid contraction of the glue, the interface is unable to deform fast enough, and acts as a drag on the glue film. This drag may be considered a back force built up in the interface as amount of deformation increases and is due to the elastic characteristics shown by any deformable body under rapid stress. Thus increasing shrinkage of the glue causes an increase in pull on the glue film itself. With the paper held taut a point is reached when the glue film fails at its weakest point, but the force required to break the glue film is greater than that required to deform the interface initially, and the breaking of the glue permits additional deformation of the interface. As shown in the highly magnified cross-sectional diagram in Figure 2, movement occurs towards the centers of the small contracting areas of glue and the cracks widen.

Once the paper is dry, this paper remains flat with changing humidities. The local areas are so small that force developed by the change in glue film can deform the interface through the necessary distance to dissipate the force, and the small deformation which is necessary proceeds sufficiently slowly not to build up any resistance.

From the foregoing it will be evident that the size of the break pattern depends upon a balance of two forces—the force developed by the shrinking glue film and the drag built up by the growing resistance of the interface to deformation. Thus, a weak, easily deformed interface or a strong, difficultly broken glue film favors a large break pattern and wide cracks. Conversely, an interface which resists deformation more strongly, or a glue film which breaks more easily, favors a small break pattern with narrow cracks. For a gummed film of given mechanical properties, the thicker the film the larger will be the break pattern formed. For an interface with given mechanical properties, the thicker the interface, the larger will be the break pattern formed.

By applying the interface without using a solvent which causes the paper to swell, the usual difficulties due to shrinkage of the paper in drying are avoided; even though the gum coating be

applied with a solvent which would ordinarily cause the paper to swell, the paper is unaffected because of the interface.

From the foregoing it will be evident that the interface is preferably formed with elastomers, that is organic substances possessing the property of forcibly retracting to approximately their original size and shape after being greatly distorted (e. g., 100% to 600%); or from plastomers, that is plastics which after being greatly distorted regain at most only a small part of their original size and shape; or mixtures of these.

It should be understood that the present disclosure is for the purpose of illustration only and that this invention includes all modifications and equivalents which fall within the scope of the appended claims.

I claim:

1. A gummed sheet of the type comprising a thin flexible cellulosic backing and a layer of non-tacky water-activatable gumming having a tendency to curl the backing by setting up contractile forces in drying, said forces being subject to variation as a result of unequal expansion or contraction of the non-tacky gumming and the backing sheet, said layer being self-broken into a multitude of small islands with gaps therebetween which separate each island from each adjacent island, and an intermediate deformable layer adhering to the backing and gumming with sufficient adhesiveness to prevent separation of the three layers in response to said variation, the intermediate layer having sufficient internal cohesion to withstand said forces and sufficient give to permit said islands to change area in response to said variation without substantially curling the sheet.

2. A gummed sheet according to claim 1 characterized in that the intermediate layer comprises an elastomeric adhesive.

3. A gummed sheet according to claim 1 characterized in that the intermediate layer comprises rubber.

4. A gummed sheet of the type comprising a thin flexible cellulose backing and a stratum of non-tacky water-activatable gumming having a tendency to curl the backing by setting up contractile forces in drying, said forces being subject to variation as a result of unequal expansion or contraction of the non-tacky gumming and the backing sheet, said stratum being self-broken into a multitude of small islands with gaps therebetween which separate each island from each adjacent island, and an intermediate adhesive stratum securing said stratum to said backing, said intermediate stratum at 25° C. having a penetration of less than 300 sec. and a deformability of more than 4% within 20 sec. under a tension of 350 kg. per sq. cm., whereby the intermediate stratum prevents separation of the gumming stratum from the backing while permitting said islands to change area in response to said variations without substantially curling the sheet.

5. A gummed sheet of the type comprising a thin flexible cellulose backing and a stratum of non-tacky water-activatable gumming having a tendency to curl the backing by setting up contractile forces in drying, said forces being subject to variation as a result of unequal expansion or contraction of the non-tacky gumming and the backing sheet, said stratum being self

broken into a multitude of small islands with gaps therebetween which separate each island from each adjacent island, and an intermediate adhesive stratum securing said stratum to said backing, said intermediate stratum having a deformability of approximately 1000% in 20 sec. at a load of 100 kg. per sq. cm., whereby the intermediate stratum prevents separation of the gumming stratum from the backing while permitting said islands to change area in response to said variations without substantially curling the sheet.

6. A gummed sheet according to claim 4 characterized in that the intermediate layer comprises an elastomeric adhesive.

7. A gummed sheet according to claim 4 characterized in that the intermediate layer comprises rubber.

8. A flat non-curling water-remoistenable gummed sheet material, comprising a thin flexible cellulosic backing, an interfere adherent thereto and to a superposed coating of a dry adhesive, characterized by being in the form of self-broken separate areas or islands, having cracks therebetween of a width greater than the expansion or swelling of such adjacent areas or islands in response to changes in atmospheric humidity.

9. Method of making dry but remoistenable gummed sheet materials which will lie flat, comprising the steps of applying to a thin flexible cellulosic backing sheet an interface, which is adherent to the backing sheet and which is characterized by exhibiting more preliminary give than the backing sheet and more subsequent resistance to deformation than the strength of the gumming layer during the drying shrinkage of the latter, applying a fluid gumming coating which is adherent to the interface, and holding the sheet taut while drying the gumming coating to cause the coating to break into a multitude of small islands which contract to widen the cracks therebetween.

10. Method of making a dry but remoistenable gummed sheet material which will lie flat, comprising the steps of applying to a thin flexible cellulosic backing sheet an interface which is at least as deformable as to have an elongation of 4% within 20 seconds at a load of 350 kilograms per square centimeter and not more deformable than to have a penetration of 300 seconds at 25° C., and which is adherent to the backing sheet, applying a fluid gumming coating which is adherent to the interface, and holding the sheet taut while drying the gumming coating to cause the coating to break into a multitude of small islands which contract to widen the cracks therebetween.

11. Method of making dry but remoistenable gummed sheet materials which will lie flat, comprising the steps of applying to a thin flexible cellulosic backing sheet an interface which is adherent to the backing sheet and which is characterized by exhibiting more preliminary give than the backing sheet and more subsequent resistance to deformation than the strength of the gumming layer during the drying shrinkage of the latter, applying a fluid gumming coating which is adherent to the interface, and drying the gumming coating while the sheet is substantially flat to cause the coating to break into a multitude of small islands which contract to widen the cracks therebetween.

CHARLES W. STILLWELL.