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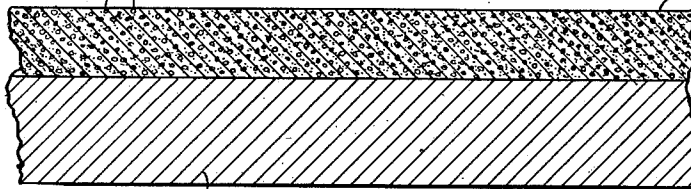
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GUMMED SHEET MATERIAL

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WATER ACTIVATABLE ADHESIVE IN CONTINUOUS PHASE
DEFORMABLE RESINOID FILLER IN DISCONTINUOUS PHASE



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GUMMED SHEET MATERIAL

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This invention relates to the art of gummed sheet material, particularly gummed paper, that is paper coated with normally dry gumming, and the problem of producing such sheet material which does not curl when drying. Since the early development of gummed paper this curling has continued to be a troublesome problem and as evidence that it has not been solved, it is a fact that up to the present time manufacturers have had to "break" the gumming after drying, as for example by drawing the paper over an edge sharply to bend the paper, thereby breaking the gum coating into very small sections along a network of cracks. This problem is particularly troublesome in the case of paper and other sheet material wetted by water because of the curling which results from forces set up when the fibers of the material are wetted and dried in the process of coating the material.

The principal object of the present invention is to produce gummed sheet material which has little or no tendency to curl when drying and which therefore need not be broken after drying. Other objects are to produce gummed sheet material which can be rendered adhesive by wetting with water, thereby avoiding the use of special moistening agents, which can be rolled or stacked sheet upon sheet without blocking, that is without superposed layers sticking together, which has quick tack when moistened with water, and which has a wide range of adhesion so that it can be satisfactorily applied to many different kinds of surfaces including moisture-proof Cellophane.

According to the present invention, the sheet material is coated on one side with a liquid mixture of water-soluble adhesive and deformable filler, with the adhesive in continuous phase and the filler in discontinuous phase, and the coating is dried, after which the gummed stock may be rolled or stacked without blocking. Preferably the coating is dried at room temperature, the paper being held flat during the drying operation. A diagrammatic illustration of the product is shown in the accompanying drawing. To stick a gummed sheet to a surface the coating is wetted with water and pressed against the surface like an ordinary gummed label, thus the dry coating is water-activatable.

The preferred water-soluble adhesive is glue or dextrin or a mixture of the two but any material or mixture of materials which becomes tacky when moistened with water may be used, such as gum arabic, some converted starches and

certain water dispersible resins such as polymethacrylic acid. The deformable filler comprises resinoids or resinous bodies such as rubber or rubber-like materials, gutta-percha or balata, polymethacrylic esters, either unmodified or modified with other materials such as a plasticizer, in contradistinction to greasy, fatty or oily materials, such as tallow. The deformable filler should be elastoplastic, comprising bodies which are elastic, that is, which have a substantial tendency to resume their original shape when deformed and which are generally compressible, or bodies which are plastic, that is, which do not have the aforesaid tendency and which are generally non-compressible, or bodies which are elastic under some conditions and plastic under other conditions, for example, which react elastically when deformed for a short time but which lose their tendency to resume their original shape when deformed for a long time. The filler should also be one which can be colloidally dispersed in water without the particles thereof undergoing any appreciable swelling.

The rate at which the aforesaid curling tendency decreases with increasing proportions of a particular filler, which may be referred to as the flatness characteristic of the filler, depends on various factors such as the size of the filler particles, the evenness of distribution of the particles throughout the continuous-phase adhesive, etc., but other factors being equal the flatness characteristics of the plastic and elastic fillers, as well as their quick-tack and range-of-adhesion characteristics, are markedly superior to those of non-deformable fillers. The deformable filler preferably comprises latex, rubber or other elastic material but may comprise a natural or synthetic resin or a resinous material to which a suitable plasticizer has been added or a mixture of two or more of any of these materials. While vulcanized latex or vulcanized rubber may be used, the filler is preferably tacky so that after the water-soluble adhesive is moistened the filler contributes to the tackiness of the moistened coating. As is well known rubber may be rendered very tacky by extensive milling or mastication and latex may be made tacky by suitable processing in liquid form. Typical ways of incorporating the filler in discontinuous phase are to emulsify latex in a glue solution or disperse rubber or a rubber solution in water and then mix with the glue solution.

For certain purposes the following tacky fillers are preferable; tacky resins such as soft poly-

basic acid-polyhydric alcohol resins (glyptals), soft Bakelites, gutta-percha resins or other natural resins with or without plasticizers, mixtures of nitrocotton with blown castor oil, soft Bakelites, etc., mixtures of chlorinated rubber with dibutyl phthalate, soft glyptals, etc., mixtures of rubber with Herculyn (the methyl ester of hydrogenated abietic acid), ethyl benzoate, decalin, etc., mixtures of normal latex with emulsions of tacky resins such as gutta-percha resin, etc. For certain other purposes the following non-tacky fillers are preferable: normal latex, unmilled rubber, synthetic rubber or rubber-like materials such as Neoprene (polymerized 2-chlorobutadiene), Thiokol (polyethylene polysulfide), Buna S (copolymer of butadiene and styrene), etc., and mixtures of film-forming materials with plastic resins in insufficient proportions to render the mixtures tacky, such as mixtures of nitrocotton with acrylate or methacrylate resins of medium or low degree of polymerization such as acryloid resins.

Typical examples of the principal kinds of deformable fillers may be classified as follows:

Tacky fillers

Plastic.—Soft alkyd resins, soft Bakelites, soft cumars, soft natural resins such as those from gutta-percha, balata, gutta siak, etc., and mixtures of these with plasticizers, mixtures of hard synthetic or natural resins with plasticizers, mixtures of nitrocotton with blown castor oil or soft Bakelites or soft alkyds, etc.

Plastic or elastic.—Alkyd resins of the type believed to be partially polymerized; e. g., Petrex A7Ht, completely blown and oxidized castor oil, soft acrylate and methacrylate resins, and gums, such as pontianick, guttas, as gutta-percha, gutta siak, balatas of high resin content, urea-formaldehyde-alkyd resin mixtures, and polymerized terpenes such as Bexin.

Elastic.—Rubber, broken down or mixed with resins, natural or wild rubber, such as Guayule, balatas of low resin content, concentrated solutions of rubber in solvents such as ethyl benzoate, decalin or other high boiling solvents, solutions of rubber plasticized with Herculyn, mixtures of latex and emulsions of tacky, elastic or plastic resins, such as gutta-percha resin emulsions, alkyd resin emulsions, gum elemi resin emulsions and rosin emulsions.

Non-tacky fillers

Plastic or elastic.—Polybutyl methacrylate resins, blown asphalts and pitches.

Elastic.—Rubber without much if any milling or processing, and synthetic rubber or rubber-like materials, such as Neoprene, Buna, Thiokol, etc.

In producing gummed sheets for application to a particular surface the water-soluble adhesive best suited to that material should be employed and other materials may be added to the glue to improve the adhesive. For example, in producing gummed labels for application to moisture-proof cellophane, the glue may first be dissolved in Trigamine (a commercial substitute for triethanolamine made by Glycol Products Co., New York city); following this, the usual dispersion with a filler is prepared. This particular mixture is useful in forming a water remoistenable adhesive which will adhere to the waxy, pyroxylin surface of moisture-proof cellophane. On the other hand the range of materials to which the gummed sheets will adhere satisfactorily may be increased by adding acids, alkalies, or organic solvents. The presence of an alkali,

such as ammonia or sodium hydroxide in the adhesive, also has a beneficial effect on certain fillers, such as latex. Typical examples of organic solvents are butyl cellosolve, ethylene chlorohydrin, methyl isobutyl ketone, diacetone alcohol and butyl lactate.

To produce a gummed sheet which has good non-curling, non-blocking and quick-tack characteristics the dry coating may comprise approximately one-quarter to three-quarters adhesive but a ratio of water-soluble adhesive to filler of approximately two to three is recommended. The following formula is particularly satisfactory because the resulting gumming not only lies substantially flat over a wide range of relative humidities, but also when remoistened, will adhere satisfactorily on an unusually wide range of surfaces of different kinds.

(1)

	Parts
(50% aqueous solution) of high viscosity dextrine, or a mixture of equal parts of high viscosity and medium or low viscosity glue	37.5
Tacky latex (50% solids) plain or vulcanized	62.5
Concentrated ammonia	2

Another preferred embodiment of the invention results from a mixture of the following compositions A and B in the ratio of 3 to 1.

(2)

A		Parts
(50% aqueous high viscosity) dextrine	95	
Butyl cellosolve (the mono ethyl ether of ethylene glycol)	5	

B

	Parts
(One-half second) nitrocotton	20
Rezyl 12-H (a glycerine phthalic anhydride modified with non-drying fatty acid)	15
Tricresyl phosphate	5
Blown castor oil	30
Ethyl acetate	30
Butyl acetate	10
Benzol	20

For gummed sheets to be applied to moisture-proof Cellophane I recommend a ratio of adhesive to filler of approximately 1 to 2, the following being particularly good formulae:

(3)

	Parts
30% solution of glue in Trigamine	50
Vulcanized latex (50% solids)	60
Concentrated ammonia	1

(4)

1 part of A mixed with 5 parts of B:

A		B	
	Parts		Parts
Glue	10	10% emulsion of milled rubber in 2% soap solution.	10
Chloroacetic acid	8		
Ethylene chlorohydrin	10		

To produce a gummed sheet which, after being moistened and applied to paper and dried, may be peeled from the paper, the ratio of water-soluble adhesive to non-tacky filler must be varied according to the nature of the paper.

For a gummed sheet to be not only flat but also peelable from hard papers I recommend a ratio

of approximately 1 to 4 as typified by the following formula:

(5)

	Parts
(50% aqueous-high viscosity) dextrine.....	6
Soft resin emulsion (50% solids).....	8
Paraffin wax emulsion (10% solids).....	15
Vulcanized latex (50% solids).....	15
Concentrated ammonia.....	1

A gummed sheet which, after being remoistened with water, adheres well to a surface similarly gummed may be prepared according to the following formula:

(6)

	Parts
(50% aqueous highly soluble) dextrine.....	15
Very tacky latex (50% solids).....	40

According to this invention the continuous phase of the coating may also comprise, as a third component, a water-soluble plasticizer such as the following: Polyhydric alcohols such as glycerol, sorbitol, mannitol; glycols; e. g., ethylene glycol, diethylene glycol, 2-methyl-2, 4 pentanediol; polyglycols, e. g., Carbowax, Igepal; di- and tri-ethanolamine; water-dispersible derivatives of above, such as glyceryl laurate, triacetate (tri-acetin), diacetate (diacetin), borate, stearate, diethylene glycol stearate, triethanolamine lactate, abietate, anhydrosorbitols and mannitols, e. g., Sorbitan, Mannitan, Mannitan laurate; salts and esters of hydroxy acids, e. g., sodium lactate, ethyl phosphate, triethyl citrate, tricarbonyl citrate; amino-alkanols, e. g., 2-amino-2-ethyl-1, 3-propanediol; ether and keto alcohols, e. g., methyl diacetone ether, benzyl ether of ethylene glycol (benzyl cellosolve); mixtures of some of the above with acids and salts such as urea, methyl urea, urethane, ammonium thiocyanate. For example, mixtures of glycerine with urea or ammonium thiocyanate plasticizer casein or glue or any other similar protein more effectively and at lower cost than glycerine alone.

In the three-component coatings the adhesive may vary between approximately one-fourth and three-fourths of the total, the filler may comprise any proportion from 0% to about 75%, that is less than approximately three-quarters of the total, and the plasticizer may comprise from 0% to about 50%, that is less than approximately one-half of the total, depending on the particular plasticizer employed and how much if any filler is employed, the proportions of plasticizer and filler varying inversely with each other. If less than approximately one-fourth of adhesive is employed, the coating does not have sufficient tack and adhesiveness, and if more than approximately three-quarters adhesive is employed the coated material tends to become substantially curly. If more than approximately three-quarters filler is employed the coating does not have quick tack; and if too much plasticizer is employed the coated material tends to block when the relative humidity exceeds approximately 65%. The range of proportions of the adhesive, plasticizer and filler is defined approximately by the triangular chart boundaries extending along the 75% adhesive line from the plasticizer base line to the filler base line, thence along the filler base line to the 50% plasticizer line, thence along the 50% plasticizer line to the 25% adhesive line, thence along the 25% adhesive line to the plasticizer base line, and thence along the plasticizer base line to the 75% adhesive line.

Thus the coated material may comprise not only 75

mixtures of adhesive and filler, as exemplified by the foregoing examples, but also mixtures of adhesive and plasticizer and mixtures of adhesive, plasticizer and filler. The following are examples of mixtures comprising plasticizer in continuous phase.

(7)

	Per cent
10 High viscosity dextrine.....	40
Sorbitan.....	25
Latex (60% solids).....	35

(8)

	Per cent
15 Medium high viscosity animal glue.....	35
Diacetin.....	40
Latex (60% solids).....	25

(9)

	Per cent
20 Low viscosity animal glue.....	55
Glyceryl monostearate.....	15
Latex (60% solids).....	30

(10)

	Per cent
25 Low viscosity animal glue.....	25
Triacetin.....	25
Latex (60% solids).....	50

(11)

	Per cent
30 Low viscosity animal glue.....	60
Methyl diacetone ether.....	30
RHoplex B-10 (solids 25% dispersion in water of methacrylate polymer).....	10

(12)

	Per cent
40 Gum arabic.....	30
High viscosity dextrine.....	15
Diethylene glycol monostearate.....	45
RHoplex WG-6 (solids) (40% dispersion in water of methacrylate-vinyl acetate copolymer).....	10

(13)

	Per cent
45 Low viscosity animal glue.....	35
Igepal CTA.....	50
RHoplex WG-6 (solids) (40% dispersion in water of methacrylate-vinyl acetate copolymer).....	15

(14)

	Per cent
55 Medium high viscosity animal glue.....	25
2-amino, 2-ethyl, 1-3 propanediol.....	10
Latex (60% solids).....	65

(15)

	Per cent
60 Medium viscosity glue.....	65
Sorbitan.....	35

(16)

	Per cent
65 Low viscosity glue.....	60
Diacetin.....	40

(17)

	Parts
Emulsion TM-8 (urea formaldehyde alkyd mixtures manufactured by Rohmand Haas).....	24.5
Sodium resinate.....	46.5
Low viscosity animal glue.....	29

From the foregoing it will be evident that each of the illustrative embodiments of the invention comprises gummed sheet material characterized

by a dry adhesive in continuous phase and a deformable filler in discontinuous phase, the adhesive comprising approximately one-fourth to three-fourths of the coating, and the product being non-adhesive and substantially non-curling when dry, whereby sheets of the material do not block when stacked together and have quick tack when wetted with water; and the remaining proportion may comprise either filler or plasticizer or a mixture of the two. By using a tacky filler large proportions of filler may be used while maintaining quick tack.

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.

This is in part a continuation of application Serial No. 220,575, filed July 21, 1938.

I claim:

1. Gummed sheet material of the water-re-moistenable type comprising a backing and on one side of the backing an exposed dry coating of water-activatable adhesive in continuous phase and a deformable resinoid filler in discontinuous phase, the ratio of adhesive to filler being approximately two to one.

2. Gummed sheet material of the water-re-moistenable type comprising a backing and on one side of the backing an exposed dry coating of water-activatable adhesive in continuous phase and a deformable resinoid filler in discontinuous phase, the ratio of adhesive to filler being approximately one to two.

3. Gummed sheet material of the water-re-moistenable type comprising a backing and on one side of the backing an exposed dry coating of water-activatable adhesive in continuous phase and a deformable resinoid filler in discontinuous phase, the ratio of adhesive to filler being approximately three to two.

4. Gummed sheet material of the water-re-moistenable type comprising a backing and on one side of the backing an exposed dry coating of water-activatable adhesive in continuous phase and a deformable resinoid filler in discontinuous phase, the adhesive comprising approximately one-fourth to three-fourths of the coating, said material being non-adhesive and substantially non-curling when dry, whereby sheets of the material do not block when stacked together and do not curl when separated from each other and have quick tack when wetted with water.

5. Gummed sheet material of the water-re-moistenable type comprising a backing and on one side of the backing an exposed dry coating of water-activatable adhesive in continuous phase and a normally tacky deformable resinoid filler in discontinuous phase, the adhesive comprising approximately one-fourth to three-fourths of the coating, said material being non-adhesive and substantially non-curling when dry, whereby sheets of the material do not block when stacked together and do not curl when separated from each other and have quick tack when wetted with water.

6. Gummed sheet material of the water-re-moistenable type comprising a paper backing and on one side of the backing an exposed dry coating of water-activatable adhesive in continuous phase and a deformable resinoid filler in discontinuous phase, the adhesive comprising approximately two-thirds of the coating, said material being non-adhesive and substantially non-curling when dry, whereby sheets of the material

do not block when stacked together and do not curl when separated from each other and have quick tack when wetted with water.

7. Gummed sheet material of the water-re-moistenable type comprising a paper backing and on one side of the backing an exposed dry coating of water-activatable adhesive in continuous phase and a normally tacky deformable resinoid filler in discontinuous phase, the adhesive comprising approximately two-thirds of the coating, said material being non-adhesive and substantially non-curling when dry, whereby sheets of the material do not block when stacked together and do not curl when separated from each other and have quick tack when wetted with water.

8. Gummed sheet material of the water-re-moistenable type comprising a paper backing and on one side of the backing an exposed dry coating of water-activatable adhesive in continuous phase and a latex filler in discontinuous phase, the adhesive comprising approximately one-fourth to three-fourths of the coating, said material being non-adhesive and substantially non-curling when dry, whereby sheets of the material do not block when stacked together and do not curl when separated from each other and have quick tack when wetted with water.

9. Gummed sheet material of the water-re-moistenable type comprising a paper backing and on one side of the backing an exposed dry coating of water-activatable adhesive in continuous phase and a normally tacky rubber filler in discontinuous phase, the adhesive comprising approximately two-thirds of the coating, said material being non-adhesive and substantially non-curling when dry, whereby sheets of the material do not block when stacked together and do not curl when separated from each other and have quick tack when wetted with water.

10. Sheet material of the character described comprising a backing and on one side of the backing an exposed coating of dry adhesive in continuous phase containing a plastic resinoid filler in discontinuous phase, the adhesive comprising approximately one-fourth to three-fourths of the coating.

11. Sheet material of the character described comprising a backing and on one side of the backing an exposed coating of dry adhesive in continuous phase containing an elastic resinoid filler in discontinuous phase, the adhesive comprising approximately one-fourth to three-fourths of the coating.

12. Sheet material of the character described comprising a backing having an exposed coating of dry adhesive in continuous phase containing a plastic filler in discontinuous phase, the adhesive comprising approximately one-fourth to three-fourths of the coating, said material being non-adhesive and substantially non-curling when dry, whereby sheets of the material do not block when stacked together and do not curl when separated from each other and have quick tack when wetted with water.

13. Sheet material of the character described comprising a backing having an exposed coating of dry adhesive in continuous phase containing an elastic filler in discontinuous phase, the adhesive comprising approximately one-fourth to three-fourths of the coating, said material being non-adhesive and substantially non-curling when dry, whereby sheets of the material do not block when stacked together and do not curl

when separated from each other and have quick tack when wetted with water.

14. A gummed sheet of the character described comprising a backing and a coating of water-remoistenable adhesive in continuous phase and, in discontinuous phase throughout the coating, a deformable resinoid filler to render the sheet substantially non-curling, said coating being hard to resist blocking and water-activatable to afford quick tack when remoistened with water, the adhesive comprising approximately one-fourth to three-fourths of the coating.

15. A gummed sheet of the water remoistenable type including a backing and, on the front of the backing, a dry coating which comprises, in continuous phase, a water-activatable adhesive which affords quick tack when remoistened with water and, in discontinuous phase, a deformable resinoid filler which renders the sheet substantially non-curling, the adhesive comprising approximately one-fourth to three-fourths of the coating.

16. Gummed sheet material of the water-remoistenable type comprising a backing and on one side of the backing an exposed dry coating of water-activatable adhesive and a water-soluble plasticizer in continuous phase and an elastoplastic resinous filler in discontinuous phase, the adhesive comprising approximately one-fourth to three-fourths of the coating, the plasticizer comprising less than approximately half of the coating and the filler comprising less than approximately three-fourths of the coating, said material being non-adhesive and substantially non-curling when dry so that sheets of the material do not block when stacked together and do not curl when separated from each other and have quick tack when wetted with water.

17. Gummed sheet material of the water-remoistenable type comprising a backing and on one side of the backing an exposed dry coating of water-activatable adhesive and a water-soluble plasticizer in continuous phase and a normally tacky elastoplastic resinous filler in discontinuous phase, the adhesive comprising approximately one-fourth to three-fourths of the coating, the plasticizer comprising less than approximately half of the coating and the filler comprising less than approximately three-fourths of the coating, said material being non-adhesive and substantially non-curling when dry so that sheets of the material do not block when stacked together and do not curl when separated from each other and have quick tack when wetted with water.

18. Gummed sheet material of the water-remoistenable type comprising a backing and on one side of the backing an exposed dry coating of water-activatable adhesive and a water-soluble plasticizer in continuous phase and a rubber filler in discontinuous phase, the adhesive comprising approximately one-fourth to three-fourths of the coating, the plasticizer comprising less than approximately half of the coating and the filler comprising less than approximately three-fourths of the coating, said material being non-adhesive and substantially non-curling when dry so that sheets of the material do not block when stacked together and do not curl when separated from each other and have quick tack when wetted with water.

19. Gummed sheet material of the water-remoistenable type comprising a backing and on one side of the backing an exposed dry coating of water-activatable adhesive and a water-sol-

uble plasticizer in continuous phase and a latex filler in discontinuous phase, the adhesive comprising approximately one-fourth to three-fourths of the coating, the plasticizer comprising less than approximately half of the coating and the filler comprising less than approximately three-fourths of the coating, said material being non-adhesive and substantially non-curling when dry so that sheets of the material do not block when stacked together and do not curl when separated from each other and have quick tack when wetted with water.

20. A gummed sheet of the character described comprising a backing and a coating of water-remoistenable adhesive and a water-soluble plasticizer in continuous phase and an elastoplastic resinous filler in discontinuous phase throughout the coating, the adhesive comprising approximately one-fourth to three-fourths of the coating, the plasticizer comprising less than approximately half of the coating and the filler comprising less than approximately three-quarters of the coating, the range of proportions of the adhesive, plasticizer and filler being defined by the triangular chart boundaries extending along the 75% adhesive line from the plasticizer base line to the filler base line, thence along the filler base line to the 50% plasticizer line, thence along the 50% plasticizer line to the 25% adhesive line, thence along the 25% adhesive line to the plasticizer base line, and thence along the plasticizer base line to the 75% adhesive line, said coating being hard to resist blocking and water-activatable to afford quick tack when remoistened with water and substantially non-curling.

21. Gummed sheet material of the water-remoistenable type comprising a paper backing and on one side of the backing an exposed dry coating of water-activatable adhesive and plasticizer in continuous phase and a normally tacky elastic resinoid filler in discontinuous phase, the adhesive comprising approximately two-thirds of the coating and the plasticizer comprising less than approximately one-half of the coating, said material being non-adhesive and substantially non-curling when dry, whereby sheets of the material do not block when stacked together and do not curl when separated from each other and have quick tack when wetted with water.

22. Sheet material of the character described comprising a backing and on one side of the backing an exposed coating of dry adhesive and plasticizer in continuous phase containing an elastic resinoid filler in discontinuous phase, the adhesive comprising approximately one-fourth to three-fourths of the coating and the plasticizer comprising less than approximately one-half of the coating.

23. Gummed sheet material of the water-remoistenable type comprising a paper backing and on one side of the backing an exposed dry coating of water-activatable adhesive and a water-soluble plasticizer in continuous phase and a latex filler in discontinuous phase, the adhesive comprising approximately one-fourth to three-fourths of the coating and the plasticizer comprising less than approximately one-half of the coating, said material being non-adhesive and substantially non-curling when dry, whereby sheets of the material do not block when stacked together and do not curl when separated from each other and have quick tack when wetted with water.

24. Sheet material of the character described

comprising a backing having an exposed coating of dry adhesive and plasticizer in continuous phase containing an elastic filler in discontinuous phase, the adhesive comprising approximately one-fourth to three-fourths of the coating and the plasticizer comprising less than approximately one-half of the coating, said mate-

5 rial being non-adhesive and substantially non-curling when dry, whereby sheets of the material do not block when stacked together and do not curl when separated from each other and have quick tack when wetted with water.

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