This invention relates to products of which gummed tape and adhesive labels are familiar examples wherein a carrier or backing of paper or like sheet material has adherent thereto a dry film of water-activatable adhesive material. On the application of the water to the adhesive surface these become usefully tacky and we may, for instance, seal a cartoon with gummed tape or apply an identifying label to an article or secure the flap of an envelope, according to the nature of the product in question. Rihthero these products have been made by preparing a water solution of the adhesive, coating it onto the carrier and drying it.

The adhesives most commonly employed for this purpose have been animal glue and starch products, such as dentinums. Gum arabic, polyvinyl alcohol and other materials which become usefully tacky when wet with water have also been employed. In the title of this specification and in the claims such water-activatable materials are for simplicity referred to as “gum.” This corresponds to the popular usage.

In referring to a film of adhesive, reference is made to the thin layer characteristic of such products at most a few thousandths of an inch in thickness. Thickness is usually expressed in terms of pounds of adhesive per ream of paper. As will appear, the films of the present invention, when comparable in linear thickness to those of the prior art, are less dense, that is, of lesser weight per ream.

The present invention provides an article having thereon a gum film which is minutely porous and which, although megascopically (to the naked eye) homogeneous, includes therein a multiplicity of air spaces of microscopic dimension so that the film may be two or three times the linear thickness of a coating of the same adhesive applied as in the prior art and having the same ream weight. The film has been referred to as being megascopically homogeneous. Lexicographically “megascopically” signifies observable with the naked eye or with a small pocket lens as distinguished from observable with a microscope. The coating contemplated by this invention appears uniform throughout its area and homogeneous when carefully observed with the naked eye as if it were a single substance of uniform structural character, the characteristic minute porosity being not directly visually observed. However, it will be understood that in commercial practice there may be imperfections, larger voids or air bubbles, perhaps in considerable number and in more or less uniform distribution which the naked eye could observe and which should not be considered as altering the essentially uniform and homogeneous character of the coating with its microscopic structure.

Articles embodying such a film coat have marked practical advantages, among which may be cited:

First: Owing presumably to the fine capillary nature of the air spaces, when water is applied to render the surface tacky it is moved rapidly and more deeply absorbed. Coatings prepared in accordance with the invention not only become tacky quicker owing to faster absorption of water but remain tacky longer because the film is more deeply penetrated by the water than those of the prior art which are relatively dense and slowly absorbent.

Second: Because of the greater thickness of the film per unit of weight, a relatively light coating will effectively cover a normally rough and fibrous surface such as that of paper. It has been found that when gum is applied to a given base material as a film having the structure characteristic of the invention, a much smaller weight of gum per unit of area than in the case of prior art films of the same adhesive is required to yield an equally good quality gum coat.

Third: The novel films exemplifying the invention have a distinctive and attractive appearance not found in the gum coats of the prior art. Even when rather dark adhesives are utilized, they are white. They are opaque, a relatively light coating normally hiding completely the backing surface or carrier to which they are applied. Generally they are glossy and, although minutely porous as described, appear megascopically as continuous unbroken homogeneous films.

The nature of the film may be further understood by a description of a novel method which forms a branch of the present invention and by which it may be prepared. This method will herein first be generally described and thereafter illustrated by numerous examples.

An adhesive solution of coatable consistency such as might have been employed in the prior art coating practice, is first prepared. There is then added thereto and emulsified therewith a substantial amount of a volatile liquid ingredient which is substantially a non-solvent of the adhesive material and substantially insoluble in the water employed in the adhesive solution. This liquid may desirably, but not necessarily, be less volatile than water. The emulsion, prepared with the assistance of a suitable emulsifying agent
if that is desirable, is of the oil-in-water type, the aqueous adhesive solution constituting the continuous phase, the non-solvent liquid the dispersed phase. The emulsion so prepared is coated on a carrier sheet in accordance with the practices of the prior art to form a thin film thereon and is then dried.

It is convenient to consider this drying as effectuated in two stages: successive in time, in one of which the water is evaporated and in the other the dispersed liquid, although a complete separation of the evaporation of the two liquids does not, or at least need not, literally occur. However, as will be described more in detail hereafter, there is a preferential initial evaporation of the water so that the continuous phase of the emulsion becomes more or less completely set and form-retaining. A certain part of the volatile liquid non-solvent will be evaporated with the water but the greater part of this non-solvent remains after the greater portion of the water has been removed. This may be considered the first stage of the drying process. In the second stage the volatile liquid non-solvent is evaporated altogether with whatever water may have remained unvaporized. During this stage the adhesive film will gradually become opaque and white, which may be attributed to the replacement as the evaporation proceeds of liquid by air in the spaces formerly occupied by the dispersed particles of liquid non-solvent. During the first stage of the drying process the adhesive solution has set to such a consistency that it is resistant to collapse during this disengagement of the non-solvent and remains in substantially the same spatial disposition as the original continuous phase of the emulsion, which remains substantially reproduced in solid form and containing voids corresponding to the original dispersed phase of the emulsion. The dried film is cellular or spongy in structure although the latter word is not here used to imply softness. It may be considered an artificially prepared solid foam. This, however, is not a statement that it is a solidified foam since the characteristic cellular structure is not formed by gas pressure and the cell walls have assumed their substantially fixed structural relationship in space before a gas replaces the dispersed liquid in the internal spaces defined by the walls.

Reference has been made to the spatial disposition of the original dispersed phase of the emulsion. While the emulsion is liquid, obviously the various particles are free to move relatively in space and it is not to be understood that a given volume of emulsion in the liquid form has a fixed and unchangeable spatial relationship of its components. Its general character is, however, substantially uniform and after it is laid down as a coating we may consider that the liquids have a substantially fixed position in space at that particular moment exhibiting the characteristics of the emulsion state. As the gum gels and as the drying proceeds, the momentary spatial disposition of the dispersed liquid becomes more set or fixed. It may be assumed that as drying proceeds the structure of the continuous phase may be more or less altered as it solidifies. Perhaps the cells are expanded by the vapor pressure of the dispersed liquid as the drying proceeds, and owing to the gels in two stages dispersed droplets will not remain completely separate but in some stage of the drying process vents to the exterior will be formed, but without collapse of the cellular structure of the external phase. There may be some tearing or rupture of the solidified or partly solidified structure of the adhesive, and, as it becomes drier, the material will shrink, losing volume in marked degree, and the cell walls will be distorted. The gel structure of the gum may in itself be considerably altered but the characteristic structure of the emulsion state, exhibiting minute droplets dispersed throughout the volume of the continuous (gum) phase, is maintained, the spaces originally occupied by these droplets appearing in the solid mass similarly uniformly distributed throughout its volume although doubtless much altered in form and in general no longer completely segregated. In this sense we may say that the resultant film may be considered as reproducing in solid form in substantial degree the original continuous liquid phase. The concept of the minutely porous gum film and the demonstration that it may practically be produced in the thin film form required without collapse on the one hand or essential disruption on the other to provide an essentially continuous and uniform, although minutely porous, layer are fundamental to the present invention.

There will now be illustrated by examples various practical forms of the invention and first will be given a series of examples illustrative of the variation in proportions of the constituents of the emulsion, the actual constituents being the same.

**Example 1a**

250 parts by weight, 70 gram bone glue, 40% solution
50 parts by weight, Solvesso Xylool

**Example 1b**

250 parts by weight, 70 gram bone glue, 40% solution
150 parts by weight, Solvesso Xylool

**Example 1c**

250 parts by weight, 70 gram bone glue, 40% solution
175 parts by weight, Solvesso Xylool

**Example 1d**

500 parts by weight, 70 gram bone glue, 20% solution
1000 parts by weight, Solvesso Xylool

The designation of the glue as "70 gram" is a commercial designation and refers to jelly strength as determined on the Bloom gonometer. Solvesso Xylool is the commercial name of a solvent understood to be essentially a mixture of several forms of di-methyl benzene (xylene) and is a hydrocarbon having a specific gravity of 0.862, a flash point of 80 degrees F., a boiling point range of 275 to 292 degrees F. and an aromatic content of at least 95%.

The ingredients set forth in the examples were prepared by mixing with 100 parts of the glue sufficient water to yield the concentration indicated, permitting it to stand for one hour at room temperature and then heating it to 140° F. until a smooth homogeneous glue solution resulted. This will be recognized as corresponding to the usual process for preparing hot glue. The resultant product might be considered a material such as would be used in accordance with the practices of the prior art for direct coating onto the carrier sheet. While at this temperature of 140° F., Solvesso Xylool, also heated to 140° F., was slowly added under vigorous agitation which was continued until a satisfactory emulsion was formed, approximately for fifteen minutes. In
the proportions indicated an emulsion of the Solvesso Xyrol in the glue solution could be expected without the aid of added emulsifying agents.

The emulsions thus prepared were then coated in accordance with the usual practice of the art on felt paper of 60 pounds basis weight, 24 x 36—600. No specific thickness of coating is obligatory. In practice the choice of thickness would be determined by the desired characteristics of the particular product having in mind the novel properties realized by the present invention. Attention is again directed to this specification. The statements hereinafter made with respect to the embodiments of the various examples are predicated on coatings varying from five to fifteen pounds per 5 ream. As will appear, a film of adhesive exemplary of the invention and weighing ten pounds per 5 ream may be equivalent in linear thickness to a prior art coating of twenty or thirty or more pounds. The coatings were dried under room conditions from ten to thirty minutes 10 during which most of the water evaporated. They were then brought into contact with a steamed drum heated at approximately 220° F. until the coatings acquired a white, glossy, opaque appearance indicative of the evaporation of the Solvesso Xyrol and its replacement by air. Such independent steps, however, are not necessary to effect the effectively sequential evaporation of the water and xylo. When similar coatings were dried at 140° F. the bulk of the water was evaporated first with consequent solidification of the film and subsequent evaporation of the xylol, as indicated by the change in the white glossy state. Other specimens subjected only to room temperature reached eventually this same condition. This illustrates the fact that, provided most of the water is evaporated prior to substantial evaporation of the dispersed liquid so that the continuous phase becomes solidified and substantially shape-retaining prior to the evaporation of the bulk of the dispersed liquid, a considerable latitude may be permitted in the conditions of drying. The drying should, of course, be carried out at film temperatures below the boiling temperatures of either the water or the dispersed liquid to avoid emulsion which would disrupt markedly the spacial disposition of the continuous phase either in its liquid or more or less completely solidified form.

The opacity developed during the drying indicates, as already stated, the fact that the dried film includes a multiplicity of microscopic air spaces. The cellular or porous structure of the dried film is also shown by the fact that it is of substantially lesser density than a film of the same linear iickness which might be coated from the original adhesive solution alone. That is, the film in accordance with the invention may be said to be expanded. The degree of expansion will vary with the amount of non-solvent liquid employed relative to the amount of dry adhesive material employed. Thus, with 50 parts of Solvesso Xyrol and 100 parts of dry glue, Example 1a, the expansion is roughly 50% to 75%. That is, the density has decreased about 33% to 43%, whereas a decrease in density of 20%, expansion by one fourth, would certainly be considered substantial. With 175 parts of Solvesso Xyrol and 100 parts of dry glue, Example 1c, the expansion is roughly 25%, corresponding to a decrease in density of 80%. In all these instances the films remain megascopically uniform and continuous and are of such character as to be practically useful. The fineness of the emulsification and the conditions of drying are factors which influence the degree of expansion. The next series of examples illustrates compositions prepared similarly to the examples of Series 1, differing only in that a glue of considerably higher viscosity is used.

**Example 2a**

250 parts by weight, 210 gram bone glue, 40% solution

100 parts by weight, Solvesso Xyrol.

**Example 2b**

280 parts by weight, 210 gram bone glue, 40% solution

175 parts by weight, Solvesso Xyrol

The next series of examples illustrates the use of a glue, probably of higher grease content, with which it is advantageous to employ a dispersing assistant or emulsifier. Many emulsifiers are known and the selection of a suitable one would present no difficulties to those skilled in the art. In the present instance a commercial product known as Aerosol OT was used. This is an ester of a sulfonated bicearboxylic acid. This material was dissolved in the Solvesso Xyrol before proceeding to emulsify although it might alternatively have been dissolved in the glue solution. Except for the employment of this agent, the examples of Series 3 were prepared in the same general manner as those of Series 1 and 2.

**Example 3a**

333 parts by weight, 250 gram bone glue, 30% solution

100 parts by weight, Solvesso Xyrol

5 parts by weight, Aerosol OT

**Example 3b**

333 parts by weight, 250 gram bone glue, 30% solution

200 parts by weight, Solvesso Xyrol

5 parts by weight, Aerosol OT

**Example 3c**

333 parts by weight, 250 gram bone glue, 30% solution

300 parts by weight, Solvesso Xyrol

5 parts by weight, Aerosol OT

**Example 3d**

333 parts by weight, 250 gram bone glue, 30% solution

400 parts by weight, Solvesso Xyrol

5 parts by weight, Aerosol OT

**Example 3e**

333 parts by weight, 250 gram bone glue, 30% solution

500 parts by weight, Solvesso Xyrol

5 parts by weight, Aerosol OT

In describing all the examples hitherto given reference has been made to a temperature of 140° F. at which the glue was dissolved and at which the emulsification was carried out. This temperature was employed to maintain the glue and the emulsified composition at a suitable fluidity.
The temperature may be widely varied providing only that a fluidity suitable for carrying on the emulsification and the subsequent coating operation is maintained. Also the amount of water used with the adhesive may be rather widely varied in a given composition, the more concentrated form requiring the higher temperature.

Many liquids other than Solvesso Xylol, referred to in the examples given, may be employed if of appropriate volatility and substantially insoluble in water and substantially non-solvents of the glue. The following is a list of a number of volatile liquids which have been determined by test as suitable. This may serve as a helpful guide in choosing other liquids.

Methyl hexyl ketone. B. P. 172.5° C. Insol. in water.

Diphenyl ether. B. P. 259° C. V. sl. sol. in water.

O-dichlor-benzene. B. P. 179° C. Insol. in water.

Solvent naphtha #3. Boiling range 60°-101° C. Insol. in water.

Solvent naphtha #6. Boiling range 89°-132° C. Insol. in water.

Butyl acetate. B. P. 118° C. Solubility (0.6/100 water, 25° C).

Cyclohexanol. B. P. 160° C. Solubility (3.6/100 water, 20° C).

Carbon tetrachloride. B. P. 76.8° C. Solubility (0.6/100 water, 20° C).

Varso #2, a petroleum solvent having a boiling range of about 305°-397° F. and a flash point of about 110°-114° F. Insol. in water.

On the other hand, the following liquids were found unsatisfactory, which may be ascribed to their greater solubility in water:

Ethyl acetate. B. P. 77.1° C. Solubility (8.5/100 water, 15° C).

Isobutyl alcohol. B. P. 107.8° C. Solubility (0.6/100 water, 20° C).

Sec. butyl alcohol. B. P. 99.5° C. Solubility (12.5/100 water, 20° C).

It will be noted that carbon tetrachloride, which has been mentioned above, has a volatility substantially greater than water. Nevertheless, as stated, it could be usefully employed and the bulk of the water could be evaporated from the continuous phase of the emulsion prior to the evaporation of the bulk of the carbon tetrachloride. It may be confidently assumed that this is due to the fact that the dispersed particles of solvent liquid are surrounded by a substantially continuous body of water-dissolved glue from which the water may be quite readily evaporated but which prevents ready evaporation of the non-solvent liquid even when the non-solvent liquid is inherently more volatile.

The examples already given have employed bone glue as an adhesive. Other grades of bone glue and various other types of glue may be used, nor is the invention limited to animal glues. It is believed that any adhesive which may be dissolved and rendered tacky with water in such a manner that it could successfully be used in preparing a water-sensitive gummed product such as gummed tape or a gummed label may be prepared in the expanded and porous form characteristic of the present invention although for various reasons or commercial considerations some may be less desirable than others. To illustrate this point examples are given of a wide diversity of adhesives which it has been demonstrated may be successfully employed in the practice of the invention.

Example 4

300 parts by weight, partially converted starch, 33% solution (continuous phase)

175 parts by weight, Solvesso Xylol (dispersed phase)

20 parts by weight, alkali lignin, 50% solution (emulsifier)

The partially converted starch referred to is a starch dextrinized sufficiently to be rendered flowable at elevated temperatures at the concentration employed but which is highly viscous. The alkali lignin employed as a dispersing agent was dissolved in the starch solution since it is insoluble in the xylol. The starch solution was prepared at 190° F, then cooled to 140° F, the non-solvent emulsified therewith and the operations carried on in the manner of the examples given in Series 1, 2 and 3. The dried film prepared from this composition was cream white (due to the brown color of alkali lignin) but opaque and glossy and in physical form similar to the products of the examples of Series 1, 2 and 3.

Example 5

222 parts by weight, dextrin, 45% solution (continuous phase)

200 parts by weight, Solvesso Xylol (dispersed phase)

10 parts by weight, Aerosol OT (emulsifier)

The dextrin employed in this example was a type commonly employed for label adhesives and was greatly lower in viscosity than the partially converted starch of Example 4. The composition was prepared in a manner similar to that of the examples of Series 3. Due to the inherent properties of the dextrin employed, the dry films exhibited a fine crazing and had greater friability than was observed from examples prepared from tougher adhesives. Otherwise the products were similar to those of Examples 1, 2 and 3.

Example 6

400 parts by weight, Elvanol 51A-05, 25% solution (continuous phase)

360 parts by weight, Solvesso Xylol (dispersed phase)

3.6 parts by weight, Aerosol OT (emulsifier)

Elvanol 51A-05 is a commercial name designating a low viscosity, cold water soluble polyvinyl alcohol. The composition was prepared in a manner similar to those in Example Series 3. The dispersed liquid was dissipated from the film on drying much more slowly than in the case of the materials of the examples previously given. The ultimate coated and dried product, however, was similar to that of the other examples and was white, opaque and glossy.

Example 7

667 parts by weight, Rhotex size, 15% solution (continuous phase)

200 parts by weight, Varsol #2 (dispersed phase)

10 parts by weight, Indoil Sulfonate B, purified (emulsifier)

Rhotex size is a commercial name for a paper size which is understood to be a polymerized sodium acrylate. Varsol #2 has been previously identified in the list of liquids useful for dispersion. Indoil Sulfonate B, purified, is a petroleum sulfonate.

The composition of this present example was
prepared in a manner similar to that of Example Series 3. Due, however, to the inherent properties of the adhesive employed, the resultant product had a lower degree of tack than in the case of the products of Example Series 1, 2 and 3. That is, this example illustrates a less effective adhesive and a resultant less effective adhesive product. The finish, presumably owing to the nature of the adhesive material, was relatively dull but the product otherwise was similar to the products of Example Series 1, 2 and 3.

**Example 8**

883 parts by weight, Methocel, 11.3% solution (continuous phase)
360 parts by weight, Varsol #2 (dispersed phase)
19 parts by weight, Indol Sulfonate T-16 (emulsifier)

The Methocel here employed is methyl cellulose of 25cps. viscosity. Indol Sulfonate T-16 is a petroleum sulfonate.

This composition was prepared at a temperature of approximately 65° F., otherwise in a manner similar to that of Example Series 3.

This composition was more difficult to emulsify than those of prior examples. The coated and dried product was white, opaque and semi-glossy.

**Example 9**

286 parts by weight, gum arabic, 35% solution (continuous phase)
200 parts by weight, Solvesso Xylo(S) (dispersed phase)
20 parts by weight, alkali lignin, 50% solution (emulsifier)

Alkali lignin was dissolved in the gum arabic solution and the composition subsequently prepared in a manner similar to that of Example Series 1, 2 and 3.

The coated and dried product of this example was somewhat friable, rather similar in this respect to the product of Example 5. In appearance it was white, opaque and glossy.

It will be apparent that the invention may be embodied in other specific forms without departing from the spirit or essential attributes thereof, and the present embodiment should therefore be considered in all respects as illustrative and not restrictive, as is in fact clear in several matters from the description itself. Reference is to be had to the appended claims to indicate those principles of the invention exemplified by the particular embodiment described and which it is desired to secure by Letters Patent.

We claim:

1. The method of preparing a film of watersensitive gum which comprises emulsifying in a water solution of the gum as the continuous phase a volatile liquid substantially insoluble in water and substantially a non-solvent of the gum, coating a film of the dispersion on a carrier and drying the film to set the gum and evaporate the liquid in substantial sequence in that order whereby the spacial disposition of the continuous phase is in substantial degree reproduced in solid form.

2. The method of preparing a film of watersensitive gum which comprises emulsifying in a water solution of the gum as the continuous phase a volatile liquid substantially insoluble in water and substantially a non-solvent of the gum, and less volatile than water, coating a film of the dispersion on a carrier and drying the film to set the gum and evaporate the liquid in substantial sequence in that order whereby the spacial disposition of the continuous phase is in substantial degree reproduced in solid form.

3. The method of preparing a film of watersensitive gum which comprises emulsifying in a water solution of the gum as the continuous phase a volatile liquid substantially insoluble in water and substantially a non-solvent of the gum, coating a film of the dispersion on a carrier and drying the film by evaporating first most of the water and thereafter most of the liquid.

4. An article of flexible sheet material having a film of solidified gum adherent thereto, the film having an exposed face which when moistened by water develop in the film an adhesive tackiness permitting the securing of the article in a position of use, the film having throughout its volume a three-dimensional cellular structure with cell walls of gum defining between them a multiplicity of air spaces of microscopic dimension.

5. An article of flexible sheet material having a film of solidified gum adherent thereto, the film having an exposed face which when moistened by water develops in the film an adhesive tackiness permitting the securing of the article in a position of use, the film being megascopically homogeneous and having a minute porosity characteristically manifested by its substantially lower density and increased absorptivity when compared with dried coated films of simple solutions of like gum, its opacity and its white color.

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