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AIR-LAID, FLEXIBLE, SELF-SUPPORTING SHEET

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This invention relates generally to fibrous sheets, and more particularly to tough, self-supporting, flexible, air-laid fibrous sheets. Still more particularly, the invention relates to strong, flexible sheets containing thermoplastic fibers which possess sufficient flexibility to be rolled into a tight roll. Still more particularly, the invention relates to flexible sheets having a smooth, slick surface on one face and a comparatively rough surface on the other face.

There has long been a need for tough, self-supporting yet flexible fibrous sheets which may be molded into a variety of useful shapes having excellent tensile strength and tear resistance, with little or no elongation. Such sheets must be flexible despite their self-supporting character and they must be susceptible of being decorated and ornamented to present an attractive appearance to the eye. Such tough, flexible, self-supporting sheets find use as luggage shells, lamp shades, automobile headliners, automobile door panels, and other molded products. Although prior sheet products for these uses have served well, sheets having the requisite combination of toughness, self-support, and flexibility, coupled with ease of decoration, have not been available.

It is the primary object of the present invention to supply such a product. It is a further object of the present invention to supply a process whereby there may be made fibrous sheets which are sufficiently flexible to be rolled into a tight roll while retaining the requisite toughness, tear resistance, and tensile strength.

These objects have been accomplished in a surprisingly straight-forward manner. The method of the present invention contemplates forming a fibrous mixture containing 20%–50% by weight cellulose acetate fibers having a length in the range of 1½ to 2 inches and a denier in the range of 40–60. The mixture also includes 20%–50% by weight cellulose acetate fibers having a length in the range of ½ to 2 inches and a denier in the range of 5.5–35. The mixture also includes at least 10% by weight cellulose fibers having a length in the range of ½ to 3 inches. The balance of the fibrous mixture is cellulose fibers as defined above, 5.5–35 denier cellulose acetate fibers of a length of ½ to 2 inches, or mixture thereof. To the well-blended mixture is added a liquid plasticizer for the cellulose acetate fibers in an amount of 2%–8% by weight based on the weight of the fibrous mixture. The plasticizer is thoroughly blended with the fiber mixture to achieve good dispersion and penetration of the plasticizer into the cellulose acetate fibers. The blended and plasticized fiber mixture is then fed to an air stream in order that a web of the blended fibers may be laid down to form an air-laid sheet having a weight in the range of 10–30 ounces per square yard. Additional liquid plasticizer within the amount defined above may then be sprayed or otherwise added to the side of the air-laid sheet having the higher concentration of smaller cellulose acetate fibers; this concentration phenomenon will be explained below. The air-laid sheet, measuring 1 to 3 inches in thickness is then subjected to elevated temperatures in the range of 250°–350° F. and elevated pressures in the range of 100–1000 pounds per square inch in order to form the tough, air-laid, fibrous sheet of the present invention.

The cellulose acetate fibers to be used in the method of the present invention are known items of commerce. Cellulose acetate is made by reacting the cellulose in wood pulp or cotton linters with acetic acid or acetic anhydride in the presence of a sulfuric acid catalyst. The cellulose molecule is fully acetylated, containing three acetate groups per glucose unit. The catalyst has caused degradation of the cellulose chain with the result that the cellulose acetate contains in the range of 200–300 glucose units per molecule. Much of the fully acetylated cellulose acetate is partially hydrolyzed by water to reduce the acetate content to an average of 2–2.5 acetate groups per glucose unit. This cellulose acetate is thermoplastic and is soluble in acetone. Fibers are generally made by forcing an acetone solution of the material through the orifices of a spinneret into a stream of warm air in order to evaporate the acetone. Fully acetylated material, frequently identified specifically as cellulose triacetate, is insoluble in acetone but is soluble in methylene chloride. The term "cellulose acetate fibers" as used in the present specification is intended to cover both the partially hydrolyzed material and the fully acetylated material.

The fibrous mixture to be used in making the product of the present invention must contain 20%–50% by weight of the large-denier cellulose acetate fibers having a length in the range of 1½ to 2 inches. Larger amounts of such fibers unduly stiffen the product, while smaller amounts below the minimum defined reduces the strength of the product. The denier of this fraction of the total fibers must be in the range of 40–60. The denier of a yarn is the weight in grams of 9,000 meters of the yarn. A 50 denier yarn or fiber is one of which 9,000 meters weighs 50 grams. It will be recognized by those skilled in the art of sheet making that the requirement of 40–60 denier cellulose acetate fibers is unusual in that such fibers are thicker than those normally used for making air-laid sheets. It is one of the surprising features of the present invention, however, that a combination of flexibility and toughness is not attained unless the defined portion of the total fibers meets the defined size and length limitation.

The second fibrous fraction in the total fiber mixture will also be cellulose acetate fibers, but this fraction, weighing 20%–50% by weight of the total mixture, may preferably have a shorter length than the first fraction, being in the length range of ½ to 2 inches. The denier of the second fraction must be smaller, being in the range of 5.5–35. The denier of the second fraction is also critical in order to achieve the desired classification effect to be described below. These thinner fibers help produce the high degree of fiber bonding required by virtue of the increased number of fiber-to-fiber contact points.

The mixture must also include at least 10% by weight cellulose fibers having a length in the range of ½ to 3 inches. Cellulose fibers are needed to impart additional toughness to the finished sheet and to serve as a kind of basic network for the thermoplastic fibers to adhere to. The cellulose fibers enhance the stiffness and smoothness of the final sheet. However, no more than about 30% by weight cellulosic fibers should be present in the final fibrous mixture if the sheet is to retain its strength and flexibility. The cellulosic fibers will have a length in the range of ½ to 3 inches and may be of cotton, cotton linters, sisal, hemp, kraft, sulfite, and other chemical and semi-chemical wood pulps, including thoroughly beaten wood fibers. The cellulosic fibers may be thought of as filler fibers as opposed to the cellulose acetate fibers which serve as binder fibers.

The balance of the fibrous mixture may be made of cellulose fibers as defined immediately above, the 5.5–35 denier cellulose acetate fibers as defined above, or mixtures thereof. Hence the basic fibrous mixture to be used in the product of the present invention will be 20%–50% by weight relatively large-denier cellulose acetate fibers, 20%–50% by weight relatively small-denier cellulose acetate fibers, 10%–30% by weight cellulosic fibers, any remaining balance to be supplied by any one of the classes of fibers described so long as the relative amounts for any one fraction do not rise above the stated maximum.

The total fibrous mixture is blended in order that all the fibers may be thoroughly admixed and in order that the bundles of fibers may be opened up as thoroughly as practical. The fibers may be fed into a conventional garnet machine or other device to loosen, fluff, and separate the fibers from the form in which they are shipped if such is desirable. Blending may be carried out in any suitable fiber-blending device, including the feed box of a "Rando-Webber" machine. The latter is a particulary useful device for carrying out the blending since it may then be used to deposit the air-laid web.

Prior to the laying of the web, however, a plasticizer or softener for the cellulose acetate fibers must be added. Such plasticizers are well-known in the art and include phosphate esters such as triethyl phosphate and tripropyl phosphate. Glyceryl diacetate and glyceryl triacetate may be used as may the phthalate esters such as dimethyl phthalate, dimethoxyethyl phthalate, dimethoxybutyl phthalate, and dibutyl phthalate. Additionally, such plasticizers as N-methyl pyrrolidone, triethyl citrate, dimethoxyethyl adepate, methyl phthalyl ethyl glycollate, and benzyl benzylate may be used. Certain of the complex ethers such as ethylene glycol monomethyl ether, and diethylene glycol monoethyl ether acetate may be used as plasticizers. The choice of the particular plasticizer or plasticizer mixture to be used will generally be determined by cost, availability, the odor that can be tolerated, and other such considerations. The plasticizer of choice is dimethoxyethyl phthalate.

The plasticizer is added in an amount of 2%–8% by weight based on the total weight of the fibrous mixture. Although the plasticizer to be used is a plasticizer only for the cellulose acetate fibers and not for the cellulosic fibers, tests have shown that more satisfactory results are obtained when the plasticizer is applied to the entire fibrous mixture as opposed to being applied solely to the cellulose acetate fibers. The cellulosic fibers apparently carry a homogeneous coating of the plasticizer, and this coating softens the cellulose acetate fibers, particularly at the points of contact with the cellulosic fibers during the subsequent heating and pressing operation. Sheets of decreased tensile strength result from applying the plasticizer or softener solely to the cellulose acetate fibers.

The well-blended fibrous mixture containing the plasticizer is then utilized in a Rando-Webber machine, or other device for laying down a web on a foraminous, felt-forming member. It may be desirable to age the fiber-plasticizer mixture for 24–48 hours to insure good plasticizer distribution and penetration. The term "air-laid" as used herein is in contrast to "water-laid" and defines a relatively light, open, and lofty web formed when the stream of air conveying the fibers passes through the foraminous member such as a wire. The fibers form a felt or a web on the moving surface of the foraminous member. The lightness and openness of the resulting felt can be controlled by varying such factors as speed of the foraminous member, amount of fibers fed into the moving air stream per unit time or unit volume of air, and the extent of the subsequent pressing and rolling operations. For the purposes of the present invention, the resulting web must have a weight in the range of 10–30 ounces per square yard. The loose web will have a thickness in the range of 1 to 3 inches.

When the defined fibrous mixture is used to deposit a web of the defined weight, it will be found that a classification of the cellulose acetate fibers takes place. The thicker cellulose acetate fibers concentrate directly on the surface of the foraminous member while the thinner cellulose acetate fibers concentrate on the surface of the web opposite the foraminous member. The filler fibers are generally scattered throughout the sheet but occur in diminishing amounts at the two faces. The result of such classification is that on subsequent heating and pressing of the web to be described below, the face of the sheet containing a predominance of the thinner cellulose acetate fibers is smooth and slick. The opposite face of the sheet, containing a predominance of the heavier cellulose acetate fibers, is more grainy and rough. This effect, the slick surface on one face of the sheet and the rough surface on the other face of the sheet, is a highly desirable situation for the proposed uses of this novel product. The smooth, slick surface allows printing or embossing operations or the adherence of very thin vinyl or other synthetic resin films thereto in order to form a smooth, attractive and decorative appearance to the eye. The opposite side of the sheet where the surface is roughened allows the better adherence of the fibrous sheet by means of adhesive to such backing materials as paper, cloth, metals, wood, plaster, and the like. Hence the critical selection of the proper fibrous mixture for forming the air-laid sheet along with the proper softener or plasticizer allows the production of a sheet having unexpected decorative properties on one face and unexpected mechanical properties on the other.

The smoothness of the surface containing the preponderance of thinner cellulose acetate fibers can be enhanced by holding out some of the plasticizer and then applying it to the smooth surface after the air-laid sheet has been formed. This post-plasticization is most conveniently carried out by spraying the plasticizer on the sheet. About 1%–3% by weight plasticizer based on the total weight of the fibers may conveniently be added this way prior to heating and pressing.

The web made as described above must be hot pressed to produce the product of the present invention. The pressing may be carried out between press platens, matched dies, calender rolls, or any suitable device for achieving the requisite temperatures and pressure. Pressing may be carried out in two or more stages if desired. The loose web may be partially compacted for example between calender rolls or preliminarily fused by the application of heat with little or no pressure and subsequently pressed or molded to achieve the desired product. The temperature to be used during at least the final stages of pressing will be in the range of 250°–350° F. in order that the thermoplastic cellulose acetate fibers may be fused to serve as the binder for the sheet. The plasticizer lowers the normal softening point of the cellulose acetate and enhances its flowability. The pressure will be in the range of 100–1000 pounds per square inch in order that the sheet may possess the requisite strength. The finished sheet will have a density in the range of 35–65 pounds per cubic foot, although it must be emphasized that the sheet weight of 10–30 ounces per square yard is not affected by the pressing and heating operation. The heat and pressure cause the surface having the predominance of cellulose acetate light fibers to form a slick, smooth surface, while the opposite surface containing the coarser fibers will retain its rough characteristics. The sheets will generally have a thickness in the range of 0.01 to 0.05 inch after pressing and heating.

The sheet may be pressed in flat sheets for use as automotive door panels, or the sheets may be pressed in molds to the desired shape, for example as automotive headliners. On cooling, such headliners are sufficiently flexible to be easily rolled or folded into a bundle and passed in through the window or door opening of an automobile body under construction, unrolled, and snapped into place as a headliner. Any desired decorative effects may be utilized on the slick surface. Rotogravure printing, or embossing followed by ink tinting may be carried out. Colored vinyl sheets, paints, combinations of flocks, paints, pigments, and additional fibers may be used to achieve the desired decorative effects.

The following examples illustrate several embodiments of the invention. All parts are by weight unless otherwise stated.

Example 1

A fibrous mixture was prepared containing 6.36 parts 50 denier cellulose acetate fibers measuring 1⁹⁄₁₆ inches in length, 6.82 parts 35 denier cellulose acetate fibers measuring 1⁹⁄₁₆ inches in length, 3.41 parts 5.5 denier waste cellulose acetate fibers measuring ½ inch in length, and 3.41 parts of garneted cotton fibers measuring 2 to 3 inches in length. The mixture was well-blended and there was then added 1 part dimethoxyethyl phthalate and the mixture was thoroughly blended for 30 minutes. The blended fibers were then aged for 48 hours.

An air-laid web was made from the fibrous mixture with a Rando-Webber machine. The air volume was over 1,000 cubic feet per minute through the air chamber. The web weight was 20.4 ounces per square yard.

The top surface of the web contained a predominance of the light cellulose acetate fibers while the back surface of the web, lying next to the web-forming wire, contained a predominance of the heavier cellulose acetate fibers.

The resulting lofty and open web measuring 2.0 inches in thickness was post-plasticized with 0.6 part dimethoxyethyl phthalate, applied to the short fiber bearing surface, passed through a calender at 250° F., and subjected to a pressure of 500 pounds per square inch.

After calendering, a flexible and fused sheet having one slick and smooth surface and another relatively rough surface was obtained. The sheet was tough with outstanding self-supporting strength, and yet flexible. Its tensile strength was 2,000 pounds per square inch, and the sheet could easily be rolled into a roll having an inner diameter of 3 inches. When the roll was released from confinement, the sheet snapped back into its original flat shape.

Example 2

This example was run to show the effect of the absence of 50 denier cellulose acetate fibers on the mix. Accordingly, a fibrous mixture containing 10 parts 35 denier cellulose acetate measuring 1⁹⁄₁₆ inches in length, 5 parts cellulose acetate fibers, all 5.5 denier but mixed between ½ inch and 1⁹⁄₁₆ inches, and 5 parts of cotton fibers was treated with 0.4 parts of dimethoxyethyl phthalate and the mixture was thoroughly blended. The mixture was aged for 48 hours.

The fibrous mixture was placed in a Rando-Webber machine and air-laid to form a web of 21.6 ounces per square yard. The web was post-plasticized with 0.3 part dimethoxyethyl phthalate and calendered at 250° F. Under the same conditions of heat and pressure used in Example 1. The fused sheets lacked self-supporting strength and were entirely too flexible for use as automotive headliners and door panels. They also contained an abundance of unbound fibers.

The above-described run was repeated twice, once using a total of 1 part of dimethoxyethyl phthalate and once using a total of 2 parts of dimethoxyethyl phthalate. The results were the same in that the final sheet was too flexible and contained too high a proportion of unbound fibers.

Example 3

The following two fiber formulations were prepared:

Ingredients	Parts No. 1	Parts No. 2
Cellulose acetate, 50 denier, 1⁹⁄₁₆ inches	8	4
Cellulose acetate, 5.5 denier, ½ and 1⁹⁄₁₆ inches	8	8
Cotton	4	4
Sisal	-------	4

Run No. 1 was plasticized with 0.6 part dimethoxyethyl phthalate. The deposited web weighed 22.5 ounces per square yard.

Run No. 2 was plasticized with 0.6 part dimethoxyethyl phthalate. The deposited web weighed 21.9 ounces per square yard.

Both sheets exhibited enrichment in the short, light cellulose acetate fibers on one surface and enrichment of the long, heavy cellulose acetate fibers on the other.

Both sheets were post-plasticized by adding 0.4 part dimethoxyethyl phthalate to the short fiber bearing surfaces and pressed in a calender at 250° F. at 500 pounds per square inch.

Both runs produced flexible, tough, self-supporting sheets having a slick surface on one face and a slightly roughened surface on the other face.

We claim:

1. A method for producing a tough, self-supporting, flexible, air-laid fibrous sheet having a smooth surface on one side and a relatively rough surface on the other side which comprises forming a fibrous mixture containing 20%–50% by weight cellulose acetate fibers having a length in the range of 1½ to 2 inches and a denier in the range of 40–60, 20%–50% by weight cellulose acetate fibers having a length in the range of ½ to 2 inches and a denier in the range of 5.5–35, at least 10% by weight cellulosic fibers having a length in the range of ½ to 3 inches, any remaining balance being selected from the group consisting of said cellulosic fibers, 5.5–35 denier cellulose acetate fibers, and mixtures thereof, adding to said mixture a plasticizer for said cellulose acetate fibers in an amount of 2%–8% by weight based on the total weight of said mixture, thoroughly blending said plasticizer with said mixture, laying down a web of said blended fibrous mixture from a stream of air to form a sheet having a weight in the range of 10–30 ounces per square yard, and hot pressing said web at a pressure in the range of 100–1000 pounds per square inch and a temperature in the range of 250°–350° F.

2. A method according to claim 1 wherein said 40–60 denier cellulose acetate fibers have a denier of 50.

3. A method according to claim 1 wherein said plasticizer comprises dimethoxyethyl phthalate.

4. A method according to claim 1 wherein said 40–60 denier cellulose acetate fibers are used in an amount of 40% by weight of said mixture.

5. A method according to claim 1 wherein the 40–60 denier cellulose acetate fibers and the 5.5–35 denier cellulose acetate fibers are used in the same amount.

6. A method of producing a tough, self-supporting, flexible, air-laid fibrous sheet having a smooth surface on one side and a relatively rough surface on the other side which comprises forming a fibrous mixture containing 40% by weight cellulose acetate fibers having a length of 1⁹⁄₁₆ inches and a denier of 50, 40% by weight cellulose acetate fibers having a length of ½ inch and a denier of 5.5, 20% by weight cotton fibers having a length in the range of 2 to 3 inches, adding to said mixture dimethoxyethyl phthalate in an amount of 3%–5% by weight based on the total weight of the fibers, thoroughly blending said phthalate with said mixture, forming a web of said blended fibrous mixture from a stream of air to form a sheet having a weight in the range of 10–30 ounces per square yard, adding to the side of said sheet containing a predominance of said 5.5 denier cellulose acetate fibers an additional 1%–3% by weight dimethoxyethyl phthalate based on the total weight of the fibers, and hot pressing the resulting web at a pressure in the range of 100–1000 pounds per square inch and a temperature in the range of 250°–350° F.

7. A tough, self-supporting, flexible, air-laid fibrous sheet having a fused, smooth surface on one side and a fused, relatively rough surface on the other side comprising a cellulose acetate containing fibrous mixture plasticized with 2%–8% by weight plasticizer for said cel-

lulose acetate fibers, said fibrous mixture comprising 20%–50% by weight cellulose acetate fibers having a length in the range of 1½ to 2 inches and a denier in the range of 40–60, said 40–60 denier fibers being concentrated on said roughened side, 20%–50% by weight cellulose acetate fibers having a length in the range of ½ to 2 inches and a denier in the range of 5.5–35, said 5.5–35 denier fibers being concentrated on said smooth side, at least 10% by weight cellulosic fibers having a length in the range of ½ to 3 inches, any remaining balance of the fibers being selected from the group consisting of said cellulosic fibers, 5.5–35 denier cellulose acetate fibers, and mixtures thereof.

8. A sheet according to claim **7** wherein said fibers of 40–60 denier comprise 50 denier fibers.

9. A sheet according to claim **7** wherein said fibers of 40–60 denier are used in an amount of 40% by weight of said fibrous mixture.

10. A sheet according to claim **7** containing 40% by weight 50 denier cellulose acetate fibers, 40% by weight 5.5 denier cellulose acetate fibers, and 20% by weight cotton fibers.

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