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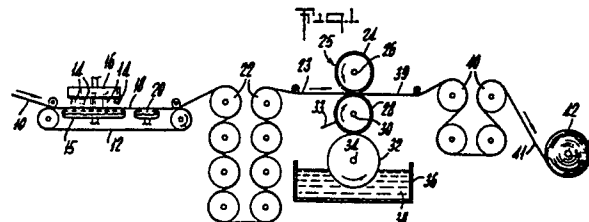
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54 Dry print bonded nonwoven fabric.

57 Entangled nonwoven fabrics are dry print bonded to produce nonwoven fabrics having an excellent combination of strength, softness, and durability.



Dry Print Bonded Nonwoven Fabric

The invention relates to a process for dry print bonding nonwoven fabrics to produce a novel nonwoven fabric product having an excellent combination of strength, softness, and durability.

Background of the Invention

10 The print bonding of nonwoven fabrics is a mature commercial technology. In a typical commercial operation, a carded or random laid web of staple-length fibers is first wetted, is optionally subjected to fluid rearrangement, is then print bonded with an aqueous resin
15 binder composition, and is then subjected to elevated temperature to dry the fibrous web and cure the binder. Early disclosures of such print bonding of nonwoven fabrics include Joshua Goldman, U.S. Patent No. 2,039,312, Esther Goldman, U.S. Patent No. 2,545,952, Drelich et al.,
20 U.S. Patent Nos. 3,009,822 and 3,009,823, and Ness et al., U.S. Patent No. 2,705,688. While the point is not addressed in most of these early patents, in commercial practice the fibrous web composed of a random array of staple-length fibers is wet when it is print bonded
25 because such a web, when dry, lacks sufficient cohesive strength to resist fiber pick off onto the print roll. (In the cited Esther Goldman patent, it is mentioned that it is preferable to wet out the web before applying binder in order to achieve better penetration of the binder.)
30 One result of printing binder onto a wet fibrous web is that the binder tends to diffuse or migrate before it cures or hardens. Because of this, a certain degree of softness, drape, and hand is lost, and harshness, stiffness, and boardiness are slightly increased.
35

One way of controlling the migration of binder is to employ binder compositions that rapidly coagulate or precipitate when deposited onto the wet web. Various ways of accomplishing this have been disclosed by Arthur Drelich
5 and coworkers, e.g., in U.S. Patent Nos. 4,084,033, 3,865,775, 3,720,562, 3,535,142, 3,536,518, and Re. 28,957. These techniques are especially useful in minimizing lateral spread of binder. Migration control techniques are preferably employed so as to have the
10 binder penetrate all the way through the web. In this respect, see Example XIII and Col. 21, lines 45 et seq. of No. 3,720,562 and Example XX and Col. 11, lines 61 et seq. of No. 3,865,775, which show the prior art position that rotogravure print bonding onto dry webs composed of a
15 random array of unentangled staple-length fibers cannot be used to produce a fabric having sufficient strength or integrity to be used commercially by itself (i.e., without being laminated to another article).

20 Until the recent past, print bonding of nonwoven fabrics has been carried out commercially mostly on carded or random laid webs, either as formed or after fluid rearrangement of the type contemplated by, for example, Kalwaites in U.S. Patent Nos. 2,862,251 and 3,033,721.
25 More recently, print bonding and/or saturation bonding has been carried out on lightly entangled nonwoven webs using fine, high pressure, columnar jets of water to lightly entangle the fibers. Such webs are first lightly
entangled and are then print bonded and/or saturation
30 bonded in one continuous operation. They are wet when the binder is applied. See Brooks, published British patent application GB 2,045,825A, November 5, 1980, and commonly assigned U.S. patent application Serial No. (CHIC-640),
filed July 13, 1981.

Russell et al., in U. S. Patent No. 3,908,058, and
Roberts, in U. S. Patent No. 3,903,342, have disclosed the
use of print bonding patterns substantially limited to the
surface, to increase the abrasion resistance of fibrous
5 webs composed at least predominantly of wood pulp fibers.

At col. 14, lines 38 et seq., of Evans, U.S. Patent No.
3,485,706, it is disclosed that water jet entangled
nonwoven fabrics may be treated with binders. It is not
10 specified whether such "treatment" is carried out on wet
or dry webs, or what type of binder pattern is used, or
how the binder is applied.

Brief Summary of the Invention

15

The invention provides a process wherein fibrous webs
composed of staple fibers are first entangled, then dried,
and then print bonded to produce novel nonwoven fabrics
having an excellent combination of softness, strength, and
20 durability.

Brief Description of the Drawings

Fig. 1 is a schematic side elevation of one form of
25 apparatus suitable for carrying out the process of the
invention;

Figs. 2-5 are photomacrographs, originally taken at 50X,
of cross-sections of nonwoven fabrics made in accordance
30 with the invention, as described in Examples 1-4,
respectively; and

Figs. 6-9 are photomacrographs, originally taken at 10X,
of the nonwoven fabrics described in Examples 1-4,
35 respectively.

Detailed Description of the Invention

Referring first to Fig. 1, a carded or random laid web 10 of staple fibers is passed onto a liquid pervious support member, such as an endless woven belt 12. The belt 12 carries the web of fibers 10 under a series of high pressure, fine, essentially columnar jets of water 14. The high pressure water is supplied from a manifold 16. The jets 14 are arranged in rows disposed transversely across the path of travel of the belt 12. Preferably, there is a vacuum means 15 pulling a vacuum of e.g., up to 5 to 10 inches of mercury, beneath the belt 12, with a vacuum slot positioned directly under each row of jets 14. The fibers in the web 10 are rearranged and entangled by the jets 14 as the liquid from the jets 14 passes through the fibrous web 10 and then through the belt 12. The fabric 18 is carried by the belt 12 over a vacuum dewatering station 20, and then proceeds to a series of drying cans 22.

Evans, in U.S. Patent No. 3,485,706, describes a process and apparatus for rearranging/entangling fibrous webs by carrying such webs on a woven belt under a series of high pressure, fine, columnar jets of liquid. Apparatus of the general type disclosed by Evans can be used in the process of this invention, although typically the degree of entanglement contemplated by this invention is less than that generally preferred by Evans.

The degree of fiber entanglement contemplated by this invention is preferably that obtained by the use of jet pressures of from about 200 to about 700 psi, and up to about 20 to 25 rows of orifices, with the orifices being spaced such that there are about 30 to 50 per linear inch. The orifices are usually about 0.005 to 0.007 inch in diameter. The web is usually positioned about 1/2 to

1-1/2 inches below the orifices. With web speeds of from about 8 to about 100 yards per minute, fibrous webs of from about 1/2 to about 5 ounces per square yard are conveniently processed.

5

The Examples below illustrate typical conditions. Selection of conditions in specific cases is dependent upon a number of interrelated factors. For instance, heavier webs usually require more energy to entangle, and therefore usually require higher pressure and/or more rows of orifices. Also, the number of rows of orifices required is directly related to the web speeds. Thus, slower web speeds (as illustrated in the Examples) require only a few rows of orifices, while faster speeds require more rows of orifices. It is well within the skill of the art to select specific entangling conditions for specific cases. As a general rule, the pressure is maintained between about 500 and 700 psi, and adjustments are made to web speed and/or number of rows of orifices to control the degree of entangling.

After the fibrous web has been entangled and dried by the drying cans 22, the dried web 23 proceeds to a rotogravure print bonding station 25 where an aqueous resin binder composition is applied to the dried web in an intermittent pattern. The dried web will ordinarily contain less than about 30 weight per cent water, based on fiber weight (30 weight per cent is about the equilibrium moisture content of a rayon web in an atmosphere having 100% relative humidity). The print bonding station 25 includes an adjustable upper rotatable back-up roll 24 mounted on a rotatable shaft 26, in adjustably controlled pressure contact with a lower rotatable engraved print roll or applicator roll 28 mounted on a rotatable shaft 30. In contact with the applicator roll 28 is a lowermost pick-up roll 32 mounted on a rotatable shaft 34. The pick-up roll

32 is partially immersed in a bath 36 of a resin binder composition 38. The pick-up roll 32 picks up resin binder composition 38 and transfers it to the applicator roll 28, which applies it to the dried fibrous web 23 as it passes
5 through the nip between the applicator roll 28 and the adjustable back-up roll 24. All the rolls are adjustable in order to be able to control the pressure at said nip. A doctor blade 33 is employed to prevent excessive build up of resin binder composition 38 on the applicator roll
10 28, i.e., to confine the binder composition 38 substantially to the grooves of the engraved pattern on the applicator roll 28 as the roll 28 contacts the web 23. As a result, the binder 38 is applied to the web 23 in an intermittent pattern corresponding to the engraving on the
15 applicator roll 28.

After the web has passed through the print bonding station 25, the printed web 39 is then subjected to elevated temperature, as by passing around a set of drying
20 cans 40, to dry or cure the resin binder, and the web 41 containing the dried or cured binder is then collected, as on a conventional wind-up 42.

The resin binder composition can be the conventional
25 aqueous latex compositions, such as acrylic latexes, polyvinyl acetate latexes, ethylene-vinyl acetate latexes, carboxylated styrene-butadiene rubber latexes, or the like.

30 The invention can use a wide variety of fibers, including rayon, polyester, nylon, polypropylene, bicomponent fibers, cotton, and the like, including mixtures thereof. Staple fibers are usually used, e.g., fibers having lengths of at least 1/2-inch and up to about three
35 inches.

The examples below illustrate the invention:

Example 1

5 A mixture of 70 weight per cent Avtex SN1913, 1.5 denier,
1-1/8 inch staple rayon and 30 weight per cent Celanese
Fortrel Type 310, 1.5 denier, 1-1/2 inch staple polyester,
was processed through an opener/blender and fed to a
random air laying unit, which deposited a $780 \pm 25\%$ grains
10 per square yard web onto a forming belt woven of 0.0157 -
inch diameter polyester monofilaments. It is a dual layer
fabric having two superimposed layers each having 42 warp
monofilaments per inch, and 32 shute monofilaments per
inch woven through the warp monofilaments in the following
15 repeating pattern: under two, between the two, over two,
between the two, etc.

Using an apparatus similar to that shown in Fig. 1, the
web was passed under a water weir to wet the fiber, and
20 was then carried at a speed of 30 feet per minute under 4
orifice strips, each of which contained a row of holes, 50
holes per inch, of 0.005 inch diameter. Water was jetted
through the holes in the orifice strips at 600 psi and
140°F.

25 The web was dewatered by passing over a vacuum slot, and
then passed over two stacks of steam cans to dry it. The
stacks of steam cans were operated at 40 psi and 80 psi
steam pressure, respectively.

30 The dried web was then run through a print station similar
to the one shown in the Fig. 1, and the following binder
formulation was printed on one side of the web:

Table 1

	<u>Component</u>	<u>Weight</u>
	Water	3.0 Pounds
5	Acrylic Resin Latex ⁽¹⁾	9.0 Pounds
	Antifoam agent (Y-30)	0.03 Pound
	Wetting agent (NS-5199)	0.21 Pound
	2 % Aqueous Hydroxyethylcellulose	2.75 Pounds
	Diammonium Phosphate	1 Gram
10	Ammonia to pH 7-8	As required
	Pigment	0.035 Pound

⁽¹⁾National Starch 4260, 51 % solids

15

The binder formulation had a viscosity of 1200 centipoises at room temperature (about 72°F.), measured by a viscometer.

20

The printing roll had an engraved pattern of straight continuous 45° diagonal lines spaced 6 lines per inch. Each line was a groove 0.004 inch deep and 0.015 inch wide. The back-up roll was rubber. The back-up roll was pressed against the printing roll by a pressure of 80 psig, i.e., sufficient pressure was used to insure that all of the binder formulation was transferred to the fibrous web. The speed through the printing station was 30 feet per minute. The printed web was then passed over two sets of steam cans set at 40 and 80 psi steam pressure, respectively.

25

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The web was collected, turned over, and print bonded on the other side by the same procedure. Total binder add-on was 5.9 weight per cent, dry solids, based on total fabric weight (average of four samples analyzed; range was 5.2 to

35

6.2 per cent). Representative properties of this fabric, and properties of the fabrics of the other examples, are displayed below in Table III.

5

Example 2

By a procedure analogous to that described in Example 1, the same base web was printed on both sides with the same printing roll. The same binder formulation was used, except that only 1.5 pounds of the hydroxyethylcellulose solution was employed. The binder formulation viscosity was therefore reduced to 280 centipoises. Total binder add-on was 7 weight per cent, dry solids, based on total fabric weight (average of four samples; range 6.6 to 7.6 per cent).

10

15

Example 3

Avtex SN1913, 1.5 denier, 1-1/8 inch staple rayon fibers were processed through an opener/blender, and fed to a random air laying unit, which deposited a $790 \pm 25\%$ grains per square yard web onto the same forming belt described in Example 1. The web was then lightly entangled by the procedure described in Example 1, except that the line speed was 36 feet per minute and the water in the jets was at 130°F.

20

25

The dried web was then printed on both sides by the following formulation:

Table II

	<u>Component</u>	<u>Weight</u>
	Water	3.0 pounds
5	NS 4260 Acrylic Latex	9.0 pounds
	Antifoam Agent (Y-30)	0.03 pounds
	Wetting Agent (NS-5199)	0.21 pound
	Diammonium Phosphate	1 gram
	2% Aqueous Hydroxyethylcellulose	2.75 pounds
10	Pigment	0.035 pound
	Ammonia to pH 7-8	As required

The viscosity of this formulation was 1200 centipoises at room temperature.

15

The printing was done by the procedure described in Example 1, except that a diamond patterned printing roll was used, and the nip between the printing roll and the rubber back-up roll was gapped by wrapping 0.007 inch thick tape around the edges of the printing roll. The diamond pattern was formed by two intersecting sets of straight continuous 45° diagonal grooves spaced 6 lines per inch. Each groove was 0.005 inch deep and 0.018 inch wide. Total binder add-on was 9.5 weight per cent, dry solids, based on total fabric weight (average of four samples; range 8.8 to 10.2 per cent).

25

Example 4

Using the same base web described in Example 3, a fabric was produced by printing both sides of the web with the binder formulation described in Example 3. The diamond pattern printing roll described in Example 3 was used, but the printing roll and the backup roll were not gapped. Total binder add-on was 15.3 weight per cent, dry solids, based on total fabric weight (average of four samples; range 14.2 to 16.7).

35

Representative physical properties of the nonwoven fabrics of Examples 1 - 4 are set forth in Table III below.

5 For comparison purposes, Table III also displays, as a
Control, typical physical properities exhibited by
nonwoven fabrics made by the same procedure described in
Example 1 except that the fabrics are wet print bonded on
one side. The webs typically contain about twice the
10 fiber weight of water when they are rotogravure printed
with binder. Although the webs are printed on one side
only, the binder penetrates to the other side.

Table III

	<u>Property</u>	<u>Ex.1</u>	<u>Ex.2</u>	<u>Ex.3</u>	<u>Ex.4</u>	<u>Control</u>
5	Softness ⁽¹⁾ , grams	24	21	40	45	58
	Wet Grab Tensile ⁽²⁾ , Pounds					
	MD	17.4	14.8	7.3	10.1	17
	CD	12.1	12.9	6.0	7.7	13
10	Elongation, %					
	MD	45	40	26	25	44
	CD	74	71	66	-	92
15	Wet Specific Grab Tenacity ⁽³⁾					
	MD	1.9	1.7	0.8	0.9	1.8
	CD	1.3	1.4	0.6	0.7	1.4
	Absorbent Capacity, % ⁽⁴⁾	836	780	740	720	680
20	Absorbent Time ⁽⁴⁾ , Sec.	2.5	2	1	1.5	3
	Wet Abrasion ⁽⁵⁾ , Cycles					
	Top Side	151	226	241	305	290
25	Bottom Side	128	227	405	517	420
	Laundryability ⁽⁶⁾ , cycles	400	400	540+	540+	100

30

(1) Standard "Handle-O-Meter" test on a 4-inch square sample using a 3/8-inch slot. Machine direction of fabric is perpendicular to slot.

35

(2) 4 x 6 inch wet sample tested in an Instron tensile tester at a pull rate of 12 inches per minute. One

gripper is 1 inch wide and the other is 1-1/2 inches wide.

(3) Wet grab tensile divided by weight in grains per
5 square yard times 100.

(4) Absorbent capacity - A five gram sample of fabric held
in a three gram wire basket is immersed in a container of
tap water. Absorbent time is the time for the sample to
10 sink. The sample is immersed for 10 more seconds, the
basket with the sample is removed and allowed to drip for
10 seconds, and is then weighed. Absorbent capacity is
calculated as follows:

$$15 \quad \frac{\text{wet weight} - \text{dry weight}}{\text{dry weight of fabric}} \times 100$$

(5) Standard abrasion test on a 3 x 9 inch sample, using a
20 5 pound head weight. "Top side" refers to the side on
which the water jets impinge; "bottom side" is adjacent
to the forming belt.

(6) Wash durability - each cycle in the wash durability
25 test is a complete agitated wash (for 10 minutes in hot
water at about 140°F. containing detergent), rinse (in
warm water - about 100°F.), and spin cycle in a Maytag
home washing machine containing an eight-pound load of
laundry. The fabric is considered to fail when it
30 develops a hole anywhere in the fabric. Two samples of
each fabric are used, with the sample size being at least
18 x 18 inches. For at least part of the wash durability
testing of the fabrics of Examples 1 - 4, an accelerated
test was used in order to save time. Instead of 10-minute
35 agitated wash cycles, 2-hour, 4-hour, and 24-hour agitated
wash cycles were used. The results reported in Table III
are the equivalent in the standard 10-minute wash cycles.

The data in Table III illustrate the unusual combination of strength, softness, and durability of the nonwoven fabrics made in accordance with the invention.

5 The beneficial combination of excellent strength, softness, and durability (as evidenced by wash durability) is believed to be a consequence of a number of cooperating factors, some of which can be seen in the photomacrographic cross-sections of the fabrics of
10 Examples 1 - 4, shown in Figs. 2 - 5, respectively. First, the softness or drapability, as measured by the Handle-O-Meter, is probably the result of the resin binder being concentrated in relatively limited spaces with an absolute minimum of diffusion or migration. In the
15 preferred mode of operation of the invention, the binder does not extend all the way through from one surface of the fabric to the other. This feature is also believed to contribute to softness or drapability. In the prior art print binding of wet fibrous webs, there is a substantial
20 amount of diffusion of binder both laterally and through the web. The diffused binder adds to stiffness or boardiness with little or no additional contribution to strength. For instance, compare the control in Table III with Examples 1 and 2.

25

The wash durability exhibited by the fabrics of this invention is little short of amazing. Again, compare the Control in Table III with Examples 1 and 2. Several factors appear to cooperate to produce this result.
30 First, the fibers are firmly embedded in the binder areas so that disentangling does not readily occur. Second, some fibers extend in the direction perpendicular to the surfaces of the fabric. Therefore, even though the center of the fabric is binder-free, it is probable that
35 virtually all of the fibers in the fabric are bonded at least twice along their lengths.

Referring now specifically to Figs. 2 - 5, cross-sections of the fabrics of Examples 1 - 4 are shown. The binder is found in discrete areas 50 with very sharp boundaries between these areas and the areas that contain no binder.

5 As can be seen in the photomacrographs, the binder is quite concentrated in the binder areas 50, and there is an absolute minimum of diffusion or migration of binder outside the binder areas 50.

10 The photomacrographs also clearly show the preferred mode of the invention wherein the binder areas 50 do not extend all the way from one surface of the fabric to the other, thereby leaving binder-free areas 52 in the center of the fabric adjacent to the binder areas 50.

15 One additional feature of the invention that can be seen in these photomacrographs is the occasional fiber 54 that extends in the direction generally perpendicular to the planes of the surfaces.

20 In order to minimize migration or diffusion of the binder so that it will be concentrated in the binder areas, and thereby achieve the optimum combination of strength, softness, and durability, the binder formulation
25 preferably has a viscosity of at least about 300 centipoises at 72°F., to about 2000 centipoises. At lower viscosities, e.g., below about 150-200 cps, significant binder migration or diffusion can begin to occur.

30 The viscosity of the aqueous resin binder compositions can be increased by adding aqueous solutions of thickeners such as hydroxyethylcellulose, acrylic acid polymers, alginates, and the like.

Typical binder solids in the binder formulation is from about 25 to about 45 weight per cent.

5 A wide variety of printing patterns can be employed. In general, the discrete binder areas should be spaced apart a distance less than the average length of the fiber used in the web, and preferably less than about one-half the length of the fiber. At the other end, the binder areas should be spaced far enough apart to maintain the
10 discreteness or separateness of the binder areas. The printing pattern can be in the form of straight lines, wavy lines, dashes, dots, annular circles ("donuts"), ovals, "torpedoes", intersecting lines (diamond pattern), and the like. The fabric can be print bonded on one side
15 only, but for optimum strength and durability is preferably printed on both sides.

The amount of binder add-on has not been found to be narrowly critical. As a general rule, the binder add-on,
20 on a dry binder solids basis, will usually be within the range of from about 1/4 to about 25 weight per cent, and preferably from about 1/2 to about 20 weight per cent, based on fiber weight.

25 Rotogravure printing is one preferred mode of carrying out the invention. However, other types of printing can be used. Examples include rotary screen printing, etc.

The novel print bonded nonwoven fabrics of the invention
30 are characterized by the following:

(a) The basic fibrous web is composed of entangled staple-length fibers. The entangling of the fibers is at least sufficient to impart to the web of entangled fibers
35 sufficient integrity to be able to subject the web, when dry and binder-free, to rotogravure printing with aqueous

binder composition with no significant picking of the fibers by the printer. (As was mentioned above, dry, unbonded staple fiber webs that are not entangled cannot be rotogravure print bonded without having individual fibers "picked" out of the web by the print roll to such a degree that fouling of the printing operation occurs in a very short time);

(b) The binder is present in the fabric in discrete areas (i.e., in an intermittent pattern) on at least one surface, and preferably both surfaces, of the fabric. The discrete areas are spaced apart a distance less than the average length of the staple fibers in the web, and preferably less than one-half the length of said fibers;

(c) The proportion of binder in the fabric is from about 1/4 to about 25 weight per cent, and preferably from about 1/2 to about 20 weight per cent, based on weight of fibers;

(d) The binder to fiber weight ratio in the binder areas per se is usually relatively high, e.g., of the order of about 1:1, binder:fiber, and higher; and

(e) Preferably, the binder areas extend through the fabric a distance less than one-half the thickness of the fabric, and more preferably, there is a binder-free region between the discrete binder areas extending from each surface.

Figs. 6 - 9 are plan view photomicrographs of the fabrics of Examples 1 - 4, respectively. The photographs were taken at exactly 10.0X to provide a convenient means for measuring the widths of the binder areas, for the purpose of determining the "spread" or increase in width over the recessed grooves in the rotogravure printing roll. Table IV, below, displays the measured widths of the binder

areas (in the 10X photographs), the actual widths, the widths of the grooves in the printing rolls, and the increase in widths.

TABLE IV

5

<u>Example</u>	<u>Binder Area</u> <u>Width, mm</u>		<u>Groove</u> <u>Width, mm</u>	<u>Increase</u>	
	<u>Measured</u>	<u>Actual</u>		<u>mm</u>	<u>%</u>
10 1	5	0.5	0.38	0.12	31
2	6	0.6	0.38	0.22	57
3	5	0.5	0.45	0.05	10
15 4	6	0.6	0.45	0.15	32

As these data illustrate, there is very little spread or area increase of the binder when it is applied to the fibrous web.

20

CLAIMS

1. Process which comprises:

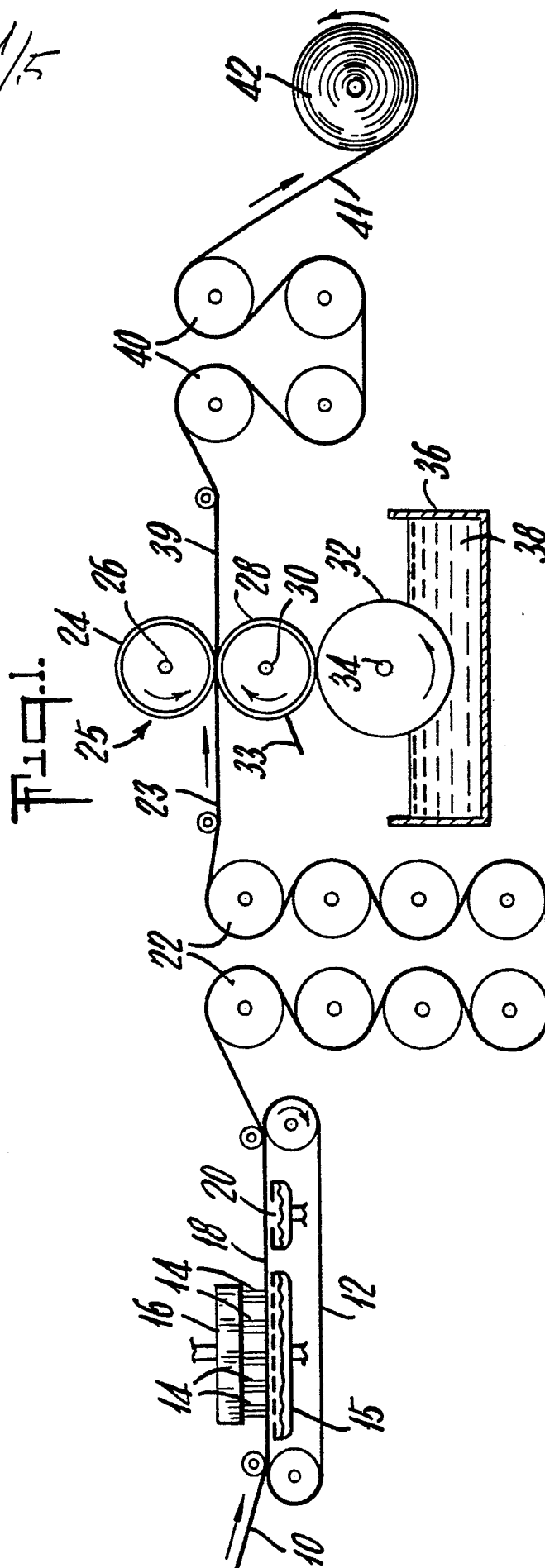
- 5 (a) supporting a layer of staple-length fibrous
starting material whose individual fibers are in
mechanical engagement with one another but which are
capable of movement under applied liquid forces, on a
liquid pervious support member adapted to move in a
10 predetermined direction;
- (b) moving the supported layer in said predetermined
direction through a zone within which streams of high
pressure, fine, essentially columnar jets of water are
projected directly onto said layer to produce a web of
15 entangled fibers;
- (c) drying the web of entangled fibers;
- (d) applying, by printing, an effective amount of an
aqueous resin binder composition to the dried web in an
intermittent pattern; and
- 20 (e) drying said aqueous resin binder composition
after it has been applied to said web.

2. Process of claim 1 wherein said binder composition is
applied to said dried web so as to produce discrete binder
25 areas that extend into said web a distance less than the
thickness of said web.

3. Process of claim 1 or 2 wherein said aqueous binder
composition is applied to both surfaces of said dried
30 web.

4. Process of claim 3 wherein said binder composition is applied to each surface of said dried web so as to produce discrete binder areas that extend into said web a distance such that a region free of binder is maintained
5 inside said web between the discrete binder areas on each surface.
5. Process of any one of claims 1 to 4 wherein said fibrous starting material is rayon or a mixture of rayon and polyester.
- 10 6. Process of any one of claims 1 to 5 wherein said aqueous resin binder composition has a viscosity of at least about 150 centipoises at 72°F.
7. Process of any one of claims 1 to 6 wherein said aqueous resin binder composition has a viscosity of from
15 about 300 to about 2000 centipoises at 72°F.
8. The bonded fibrous web produced by the process of any one of claims 1 to 7.

1/5



2/5

Fig. 2.

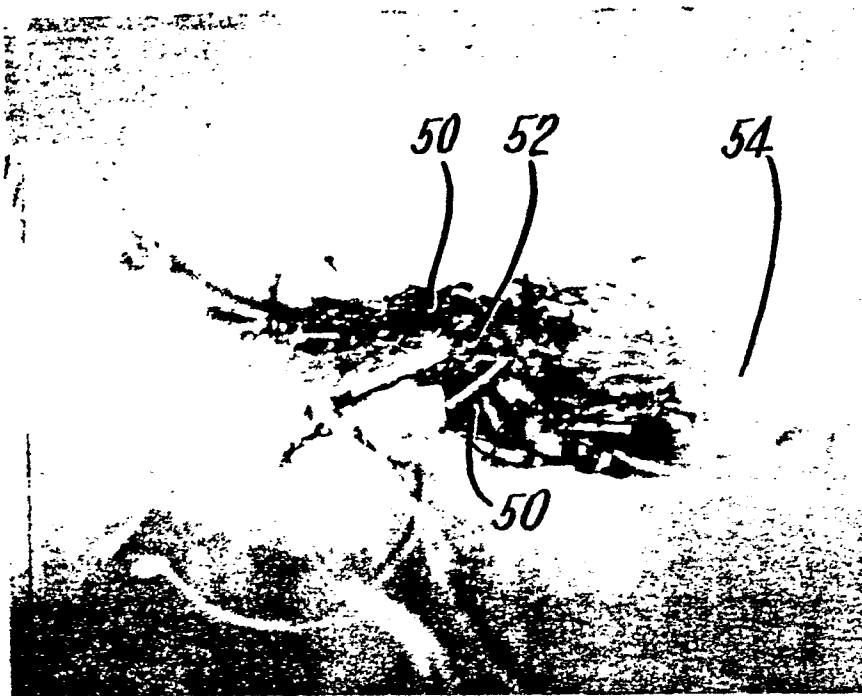
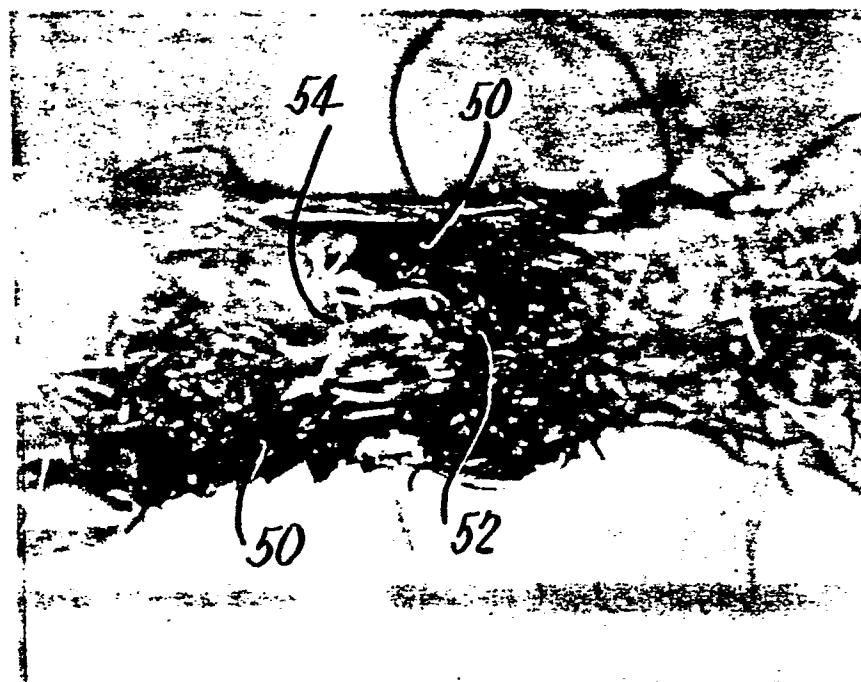


Fig. 3.



3/5 Fig. 4.

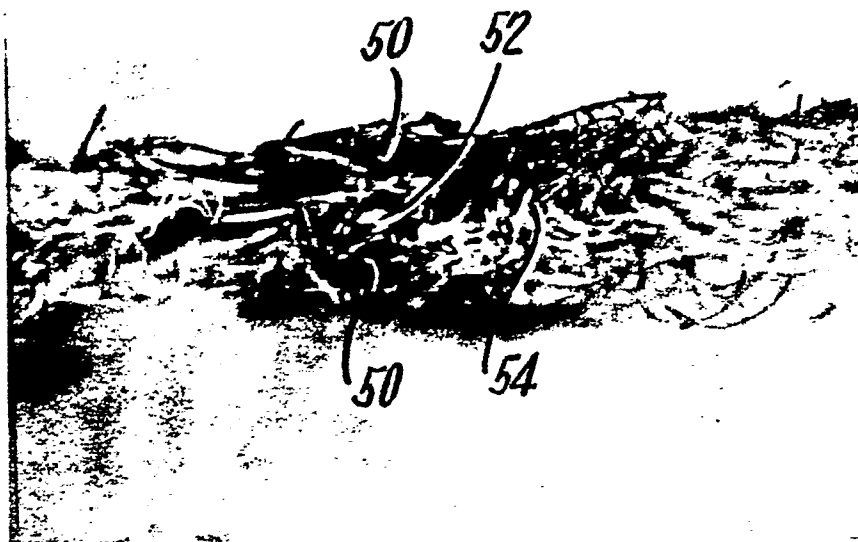
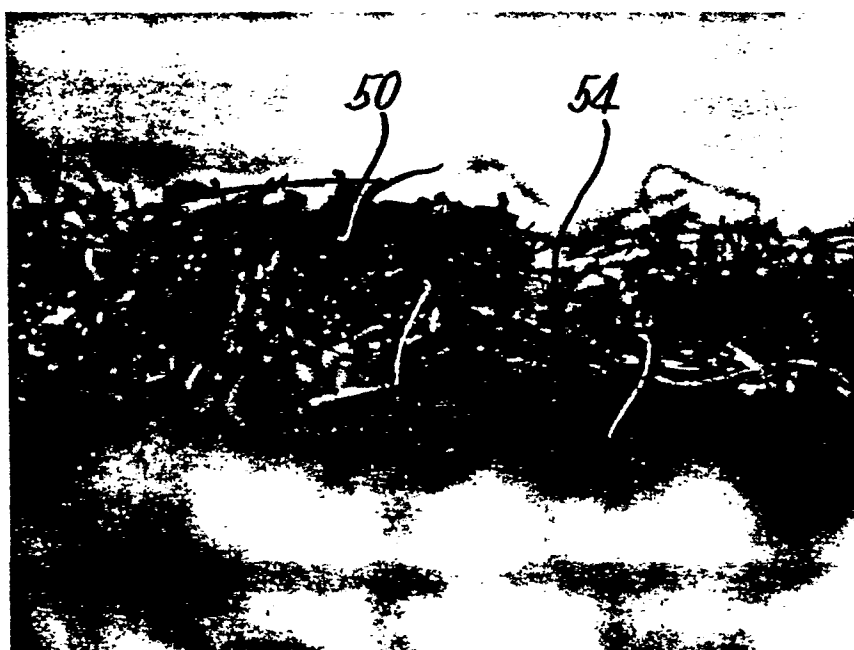


Fig. 5.



4/5 Fig. 6.

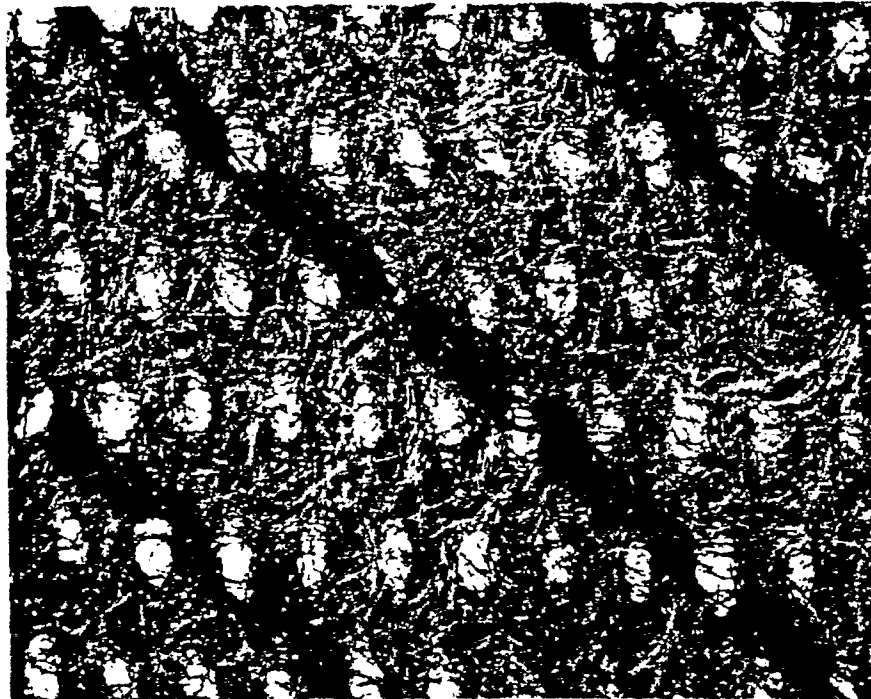
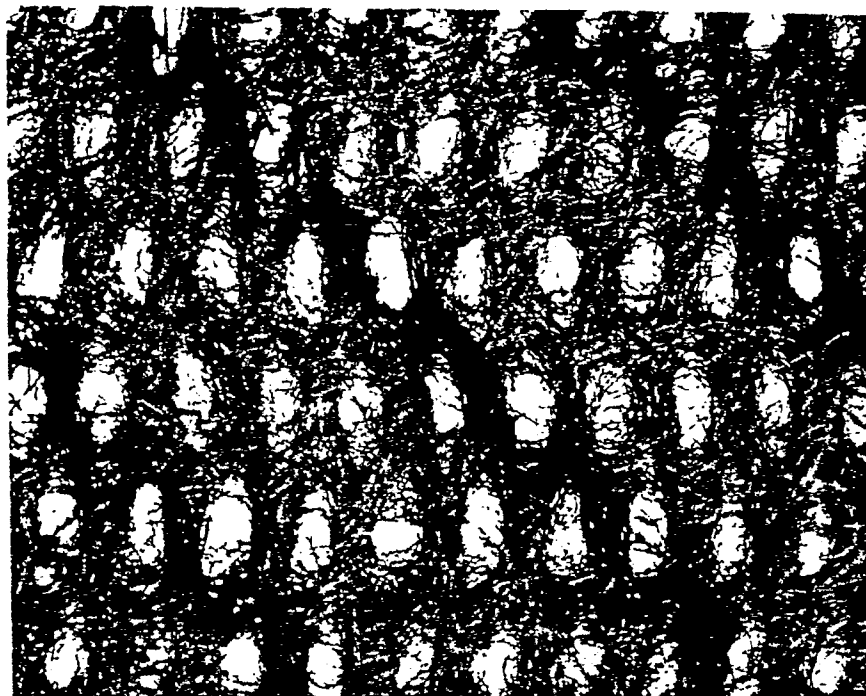
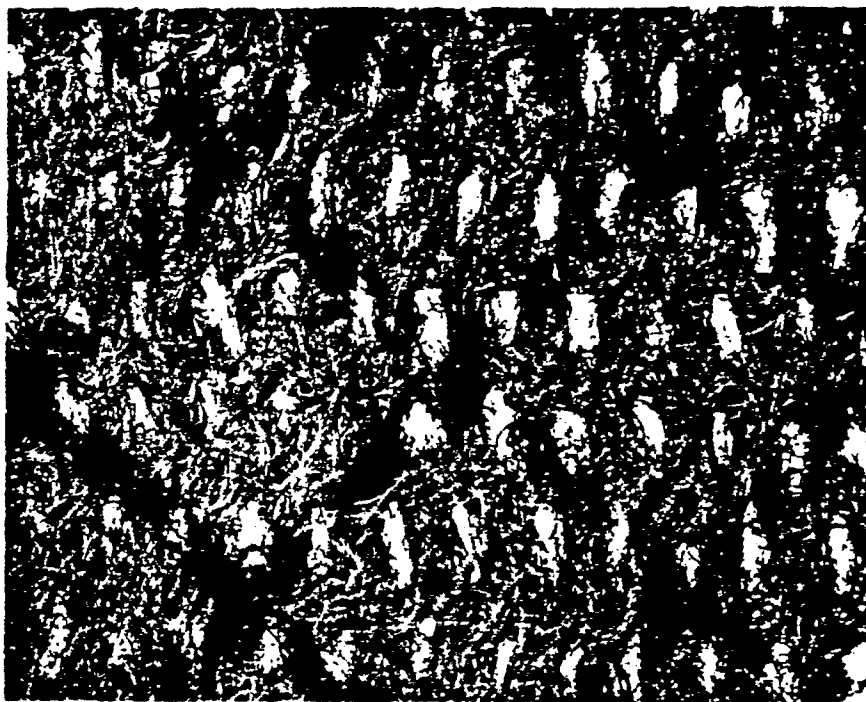


Fig. 7.



5/5 F19.8.



F19.9.

