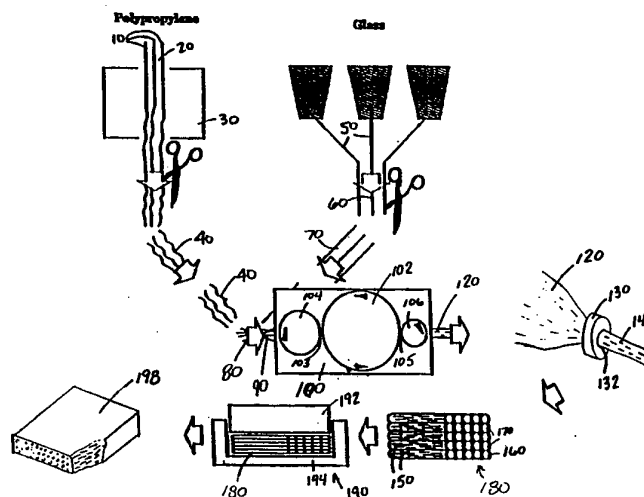




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(54) Title: COMPOSITES AND METHODS OF MANUFACTURING THE SAME



## (57) Abstract

Composites incorporating unidirectional discontinuous reinforcing fibers are formed in one process by carding a mixture including discontinuous fibers of a thermoplastic material and discontinuous fibers of a reinforcing material to form a web, collecting the web into a sliver and fusing one or more slivers to form a substantially continuous thermoplastic phase surrounding the unidirectional discontinuous fibers of the reinforcing material. In another process, the composites are formed by coextruding the thermoplastic material around substantially continuous fibers of the reinforcing material, severing the extrudate transversely to the fiber direction to form a multiplicity of pieces, juxtaposing the pieces so that the fibers in each piece extend substantially codirectionally with one another, fusing the pieces to form a unitary mass, and shearing the mass in a direction parallel to the fiber direction to redistribute the fibers in the mass. The composites in accordance with these processes have physical properties which generally are greater with respect to loads in the fiber direction than with respect to loads in directions transverse to the fiber direction.

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COMPOSITES AND METHODS OF MANUFACTURING THE SAMEBACKGROUND OF THE INVENTION

The present invention relates to the field of composite materials and processes for making the same.

Various materials are incorporated in thermoplastics to modify the properties of the base thermoplastic material. Thus, elongated fibers of reinforcing materials such as glass, metal, thermosetting polymers or high strength thermoplastic materials often are incorporated in a base thermoplastic material to form a composite having higher strength than the base thermoplastic. So called "continuous fiber" composites have reinforcing fibers which are relatively long in comparison to the overall dimensions of the composite article. Each fiber may have a length which is many thousands of times its diameter. Continuous fiber composites can be made by various processes such as hand layup or coextrusion, in which the fibers are positioned in predetermined locations within the base or matrix material. Processes for making continuous fiber composites are expensive and limited with respect to the shapes of the article which can be produced and the orientation of the fibers within the article. Moreover, continuous fiber composites incorporating very long fibers of reinforcing materials having high elastic modulus and low toughness may be relatively brittle. In such composites, any elongation of the composite results in an elongation of the fibers which is equal to or nearly equal to the elongation of the composite itself. Therefore, the fabrics will break upon relatively small elongation of the composite.

Considerable effort has been devoted towards development of so-called discontinuous fiber composites. In discontinuous fiber composites, each fiber has a length substantially smaller than the dimension of the composite article in the direction of the fiber length. Loads applied to discontinuous fiber composites are shared between the matrix and the fibers. Discontinuous fiber composites therefore can provide useful combinations of properties, such as

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combinations of relatively high strength and elongation. The orientation of the fibers in a composite strongly influences the structural properties of the composite. So-called "unidirectional" discontinuous composites have substantially all of the fibers in the entire composite, or in a substantial region of the composite, extending generally parallel to one another in a preselected fiber direction, whereas so-called "random" discontinuous composites have fibers extending in substantially random directions. Random composites have substantially isotropic properties in two or in three directions. That is, the physical properties of the composite are substantially the same in two or in three directions. By contrast, unidirectional composites generally have anisotropic physical properties. Their strength and elastic modulus generally are greater with respect to loads in the fiber direction than with respect to loads in directions transverse to the fiber direction. Generally, the physical properties of unidirectional composites in the fiber direction are superior to those of random composites. Unidirectional discontinuous composites therefore are particularly useful in structural elements intended to resist loads in a particular direction. A unidirectional discontinuous composite article typically is fabricated so that the fiber direction is parallel to the direction in which the greatest tensile loads will be applied.

The processes utilized heretofore for manufacturing discontinuous composites, and particularly those used for manufacture of unidirectional discontinuous composites, have suffered from serious drawbacks. Discontinuous composites incorporating thermoplastic-based resins have been fabricated by forming a mass of molten thermoplastic with fibers dispersed therein. For example, a mass of thermoplastic material can be subjected to an injection molding process wherein the molten mass is forced into a mold under pressure. The flow of the thermoplastic tends to orient the fibers to some degree. However, it is difficult to obtain complete orientation of the fibers in the desired direction. Also, presence of the fibers in the molten mass materially impedes flow of the thermoplastic which in turn imposes significant restrictions on the design of the mold, on the design of the article and on the process.

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One approach to fabrication of discontinuous fiber composites is described in European Patent 0.062,142. The European '142 patent suggests forming a suspension of "short cut" thermoplastic fibers and short cut reinforcement fibers in a "fluid carrier medium" and then causing that suspension to flow over a porous wall such as a porous foil or fleece. The fluid carrier medium is forced through the porous wall, whereas the fibers are retained on the porous wall. In this process, the fibers are said to be oriented substantially in alignment with one another on the porous wall. This layer of oriented fibers is then "cleansed of adhering residues of the carrier medium" and removed from the porous wall. The cleansed layer is then subjected to heat sufficient to melt the thermoplastic, thereby fusing the thermoplastic material into a coherent mass with the fibers embedded therein. Manifestly, the need to separate the oriented fibers from the fluid carrier and cleanse the oriented fibers of the carrier residue imposes undesirable process constraints. Also, the requirement to keep the thermoplastic fibers and the reinforcing fibers uniformly mixed with one another in a suspension will impose additional process constraints, particularly where the thermoplastic fibers differ significantly in specific gravity from the reinforcing fibers, as is often the case.

Accordingly, there have been substantial, unmet needs heretofore for improved processes for fabricating discontinuous fiber composites incorporating thermoplastic resins and additional materials, such as reinforcing materials, in fiber form.

#### SUMMARY OF THE INVENTION

The present invention addresses these needs.

One aspect of the present invention provides a method of making a unidirectional discontinuous composite including a thermoplastic material and an additional material in fiber form. A method according to this aspect of the invention includes the step of mixing discontinuous fibers of the thermoplastic material and discontinuous fibers of the additional material to form a mixture and then carding

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the mixture so as to orient the fibers in the mixture substantially codirectionally with one another. The carded, generally unidirectional fiber mixture is used as a preform. The preform is subjected to a fusion step in which the thermoplastic fibers are fused to form a substantially continuous thermoplastic phase surrounding the discontinuous fibers of the additional material.

The additional material preferably is a reinforcing material having a higher elastic modulus than the thermoplastic material. For example, the reinforcing material may be selected from the group consisting of glass, ceramics, metals, carbon, nonthermoplastic polymers and thermoplastic polymers having a heat distortion temperature higher than the thermoplastic material of the fibers used to form the continuous phase.

The step of forming the preform may also include the step of forming the carded fibers into an elongated intermediate preform, such as a rope-like sliver so that the codirectionally extending fibers extend generally parallel to the direction of elongation of the intermediate preform. The preform preparation step may further include the step of juxtaposing a plurality of lengths of the intermediate preform or sliver with one another so that these lengths extend generally codirectionally with one another. The fusing step may include the step of subjecting the preform to heat so as to bring the thermoplastic material in the thermoplastic fibers to a flowable condition and compacting the heated preform while maintaining the thermoplastic material in a flowable condition. For example, the preform may be squeezed between a pair of opposed members, such as the opposed portions of a compression mold. A wide variety of thermoplastic materials can be used. However, polyolefins are particularly preferred. The carding operation produces a high degree of orientation, which is retained throughout the subsequent steps of the process. The carding process is environmentally safe and does not contaminate the materials with a carrier fluid. Articles of substantially any desired dimensions can be produced readily and economically, with good quality.

A further aspect of the invention provides another process for making unidirectional discontinuous composites incorporating a thermoplastic material and an additional material in fiber form. A process according to this aspect of the invention includes the step of extruding the thermoplastic material on substantially continuous fibers of the additional material to form an extrudate with the continuous fibers extending substantially in a machine direction, i.e., the direction of extrusion. The extrudate is then severed along cutting planes transverse to the machine direction to form a multiplicity of pieces, each including relatively short fibers of the additional material together with the thermoplastic material. These pieces are then juxtaposed with one another so that the short fibers in the pieces extend substantially codirectionally with one another in a preselected fiber direction. The thermoplastic material in the juxtaposed pieces is fused to form a unitary mass including the thermoplastic material together with the fibers, the fibers still extending substantially in the fiber direction. The unitary mass is subjected to shear in a direction parallel to the fiber direction while maintaining the thermoplastic material in the mass in a flowable condition.

The shear serves to redistribute the fibers in the fiber direction. Thus, although the fibers in the mass immediately after fusion may be in substantially end to end disposition at locations corresponding to the original severing or cutting planes and the ends of the individual pieces, they are redistributed to side by side, overlapping and interleaved disposition by the applied shear. This materially enhances the physical properties of the composite.

The extruding step may include the step of coextruding the thermoplastic material with one or more strands, each including a multiplicity of fibers. This coextrusion may involve pultrusion, i.e., a process in which the fibers or strands are pulled through a die by forces supplied to the extrudate downstream of the extrusion die. The unitary mass may be subjected to shear by engaging the mass between confronting surfaces of a pair of opposed members while moving one of the surfaces relative to the other one of the surfaces

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substantially in the fiber direction. For example, the mass may be passed through a nip defined between a pair of opposed rollers while rotating the rollers at unequal surface velocities. The shearing step and the fusing step may occur concomitantly with one another. For example, the mass may be fused and sheared in a single pass through a roll mill. Substantially the same wide variety of materials can be used in this process as in the aforementioned carding and fusing process. This process provides a simple and effective way to form discontinuous unidirectional composites.

These and other objects, features and advantages of the present invention will be more readily apparent from the detailed description set forth below taken in conjunction with the accompanying drawings.

#### BRIEF DESCRIPTION OF THE DRAWINGS

A more complete appreciation of the subject matter of the present invention and the various advantages thereof can be realized by reference to the following detailed description, in which reference is made to the accompanying drawings in which:

Figure 1 is a diagrammatic view showing the process in accordance with one embodiment of the present invention; and

Figure 2 is a diagrammatic view showing the process in accordance with another embodiment of the present invention.

#### DETAILED DESCRIPTION OF THE PREFERRED EMBODIMENTS

One process according to the present invention utilizes textured strands formed from a thermoplastic material. Generally, such thermoplastic materials may comprise any organic polymer which will generally retain its shape at room temperature, but which can be deformed at elevated temperatures. Polyolefins are particularly preferred materials in that regard.

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Referring to Figure 1, such thermoplastic materials are formed into continuous fibers 10 in accordance with generally known techniques. These fibers are then collected into strands 20 consisting of a multiplicity of fibers bundled together. The number of fibers forming a strand will depend upon the particular thermoplastic material employed. For instance, when the thermoplastic material comprises polypropylene, there may be approximately 70-150 of such fibers in a strand. The strands 20 then undergo a texturing process 30 which may comprise conventional methods of crimping the strands, such as by heating the strands above their heat distortion temperature and then rolling a gear along the length of the strand, or by well-known stuffer box techniques. Alternatively, texturing may be effected by heating the strands above their heat distortion temperature and then directing a jet of air at the strands to deform same. Subsequently, the strands are cooled to retain the deformed shape.

After texturing, the continuous bundles or strands 20 of fiber are cut into a plurality of discontinuous fiber bundles 40. Preferably, the length of these discontinuous fiber bundles is between about 0.5 inches and 2.5 inches, and more preferably between about 1.50 inches and about 2.0 inches.

The discontinuous fiber bundles 40 are mixed with discontinuous fibers of a reinforcing material to form a mixture. The reinforcing materials are preferably materials having a higher elastic modulus than the thermoplastic material. Particularly preferred reinforcing materials are fibers selected from the group comprising glass, ceramics, metals, carbon, nonthermoplastic polymers and thermoplastic polymers having a heat distortion temperature higher than that of the thermoplastic material from which the fiber bundles 40 are formed. Continuous fibers 50 of these reinforcing materials, formed in accordance with conventional techniques, are collected into strands or bundles 60, each of which may include a plurality of fibers. The number of fibers in each strand will depend, to a large extent, upon the specific reinforcing material being used, and may include anywhere from two fibers to tens of thousands of fibers. In the case of fiberglass reinforcing materials, these strands will typically include

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between about 2,000 and about 4,000 glass fibers. The continuous strands 60 of these reinforcing material fibers are then cut into a plurality of discontinuous fiber bundles 70. The length of these discontinuous fiber bundles is preferably between about 0.50 inches and about 2.5 inches, and more preferably between about 1.50 and 2.0 inches. In order to facilitate the mixing and subsequent processing steps, the bundles 70 of the reinforcing material preferably have a length which is similar to the length of the thermoplastic material bundles 40.

Predetermined amounts of the discontinuous thermoplastic fiber bundles 40 and the discontinuous reinforcing material fiber bundles 70 are then introduced into a conventional precarding apparatus 80 in which the bundles 40 and 70 are at least partially unbundled so that the individual fibers therein become separated and intimately mixed with one another in a three-dimensional fashion to form a homogeneous mixture 90. In one form of precarding apparatus 80, a bed of course needles protrude from each one of a pair of confronting conveyor belts arranged to move in opposite directions. As the bundles 40 and 70 are fed into the apparatus 80, the needles separate the bundles from one another and pull the individual fibers in the bundles at least partially apart so that the fibers of the reinforcing material can become enmeshed and intimately mixed with the fibers of the thermoplastic material. As they exit precarding apparatus 80, the fibers in mixture 90 exhibit no preferred orientation, each of the thermoplastic material fibers and reinforcing material fibers being randomly arranged with respect to one another. The reinforcing material desirably constitutes between about 10 weight % and about 70 weight %, and more desirably, between about 20 weight % and about 60 weight % of the mixture.

From the precarding apparatus 80, mixture 90 is fed into a carding machine 100 in which the mixture is mechanically separated into individual fibers and formed into a cohesive web 120. The carding machine 100 includes a rotating cylinder 102 covered with a wire clothing having many fine wires protruding therefrom. A similar wire clothing covers rotatable cylinders 104 and 106. Cylinder 102

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rotates in the same direction but at a faster speed than cylinders 104 and 106. As the mixture 90 passes through the nip 103 formed between cylinder 102 and 104 and the nip 105 formed between cylinder 102 and cylinder 106, the relative movement of the wires on one cylinder with respect to the wires on the adjacent cylinder pull and tease the fibers apart. As a result of this pulling and teasing process, the individual fibers will become substantially aligned codirectionally with one another in the machine direction, i.e., transverse to the axis of rotation of the cylinders. As they are pulled apart, the fibers become entwined with one another to form a continuous thin veil or web 120 several fiber diameters in thickness. The width of web 120 will depend upon the size of carding machine 100, but will typically be on the order of about 1.0 meters. The texture of the thermoplastic fibers helps hold this web together. Carded webs of this sort typically have a bulk density which is between about 0.2% and about 0.7% of the true density of the mixture, depending on the materials carded, their relative proportions and their fiber lengths. For webs including about 70 wt% polypropylene strands and about 30 wt% fiberglass strands, web densities of between about 0.003 gm/cm<sup>3</sup> and about 0.01 gm/cm<sup>3</sup> are obtained, as compound to about 1.4 gm/cm<sup>3</sup> for a fully dense composite of this composition.

After exiting carding machine 100, web 120 is fed through a die 130 having an orifice 132. As it passes through orifice 132, web 120 is collected into a sliver 140 which will typically have a diameter of between about 2.0 cm and about 5.0 cm, and preferably will be about 4.0 cm. Thus, in a typical process, each linear meter of the one meter wide web will be collected into a linear meter of a 0.04 meter diameter sliver. The bulk density of the resultant sliver will, of course, depend upon the densities of the thermoplastic and reinforcing fibers themselves, as well as the proportion of each in the mixture 90. However, slivers consisting of about 70 wt% polypropylene and about 30 wt% fiberglass will typically have a density of about 0.005 gm/cm<sup>3</sup>. The codirectional orientation of the fibers in the web 120 will be substantially unaffected as web 120 is formed into sliver 140. Consequently, the fibers in sliver 140 will extend substantially

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codirectionally in a direction parallel to the elongation direction of the sliver.

The sliver 140 may serve as a preform for subsequent processing steps. Thus, the sliver may be cut into discrete lengths 150, a plurality of which may be arranged adjacent to one another to form a layer 160 in which the lengths 150 all extend generally codirectionally with one another. Additional layers 170 may be formed in substantially the same fashion and superposed upon layer 160 to form an unconsolidated assembly 180 in which the sliver lengths 150 in all of the layers extend in generally the same direction. The number of sliver lengths in each layer, the number of layers and the length dimension of the sliver lengths 150 will dictate the ultimate size of the mass produced after consolidation.

The assembly 180 is then subjected to a preliminary heating step to soften the thermoplastic material in the slivers. The time and temperature at which this heating step is conducted will depend to a large extent upon the particular thermoplastic material employed and the size of the assembly 180. The assembly 180 may be heated in an oven or other suitable apparatus to an oven temperature which, for polypropylene materials, is between about 200°C and about 260°C, and preferably between about 215°C and about 250°C. The duration of the heating cycle will preferably be at least about two minutes to assure that the thermoplastic material in assembly 180 is heated throughout. Heating cycles of between about four minutes and about six minutes are most preferred. For processes in which the reinforcing material is also a thermoplastic material, the heating step should be carefully controlled to assure that the thermoplastic material having the lower heat distortion temperature softens, but that the reinforcing material having the higher heat distortion temperature does not.

In order to facilitate its movement into the oven and out therefrom for further processing, assembly 180 desirably will be placed between webs (not shown) of a material which will remain thermally stable and not deform during the heating cycle. More desirably, the web material will not strongly adhere thereto after

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further processing of the assembly 180. Particularly preferred materials having these characteristics are Teflon coated fabrics.

When the assembly 180 has been heated sufficiently to soften the thermoplastic material therein to a flowable condition, the assembly 180 is compacted to form a solid, unitary mass. In a typical compacting process, the assembly 180 will be subjected to a compressive load under which the fibers of the thermoplastic material will flow together and fuse with one another to form a composite article having a substantially continuous thermoplastic phase surrounding the discontinuous fibers of the reinforcing material. The load is generally applied to assembly 180 in a direction transverse to the fiber direction so that the unidirectional orientation of the fibers therein remains substantially intact, and is then maintained for a length of time sufficient for the thermoplastic material to cool to a non-flowing state. At that point, the composite article will not distort upon removal of the compressive load. The compacting process is preferably conducted at a pressure of between about 150 atm and about 250 atm, and more preferably between about 180 atm and about 220 atm. Desirably, the compressive load is applied for between about 15 seconds and about 1.5 minutes, to assure that the thermoplastic material has completely fused together.

In preferred compacting processes, the load-applying members are shaped to produce the desired shape in the compacted article. In an example of one such compacting process, the heated assembly 180 is placed in a compression mold 190 having opposed members 192 and 194 which define substantially the final shape of the article to be formed. These opposed members are at a substantially cooler temperature than the temperature of the assembly 180. The compressive load applied to assembly 180 as the opposed members converge toward one another causes the thermoplastic material to flow together and fuse into a substantially continuous phase. The compressive load is maintained for a sufficient period of time for the thermoplastic material to cool to a non-flowing condition after which the opposed members are opened to yield a composite article 198 of the desired shape.

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By substantially following the process as described above, a unidirectional discontinuous composite is formed. The composite desirably includes a continuous phase of a thermoplastic material surrounding a plurality of discontinuous fibers of a reinforcing material oriented codirectionally with one another.

Composites formed by the above-described process have anisotropic physical properties. Thus, these composites have strengths and elastic moduli which generally are greater with respect to loads in the fiber direction than with respect to loads (in the plane of the composite) in directions transverse to the fiber direction. The magnitude of these properties will depend upon the particular thermoplastic and reinforcing materials from which the composites are fabricated. Preferred composite materials comprising discontinuous strands of fiberglass surrounded by a continuous phase of polypropylene in a ratio of about 30 wt% fiberglass and about 70 wt% polypropylene typically exhibit room temperature tensile strengths in the fiber direction which are about 2-5 times the room temperature tensile strengths in directions transverse to the fiber direction. Room temperature flexural strength values which are at least 1.5-3 times greater in the fiber direction than in directions transverse to the fiber direction are typically obtainable with preferred composites. Further, the flexural modulus of preferred composites is generally at least 1.5-4 times greater in the fiber direction than in transverse directions, as measured at room temperature.

Additionally, preferred polypropylene/fiberglass composites according to the above-described process exhibit toughness properties which are anisotropic. The typical toughness values for these composites as measured by the Charpy Impact test are also about 1.5-4 times greater in the fiber direction than in transverse directions.

In a further embodiment of the invention, thermoplastic composites incorporating unidirectional discontinuous reinforcing fibers are formed from pultruded pellets consisting of strands of a reinforcing material surrounded by a coating of a thermoplastic material. This process uses substantially the same thermoplastic

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materials and reinforcing materials as the process described above. Although this process begins with reinforcing materials which are again in the form of continuous fibers or strands, the thermoplastic material need not be in fiber form, but rather may be provided in the form of pellets, granules, flakes, powders, or any other divided form.

In the first stage 200 of this process, the thermoplastic material and reinforcing fibers are coextruded to form a continuous string 210 consisting of the continuous fiber 212 of the reinforcing material surrounded by a coating 214 of the thermoplastic material. This coextrusion step may comprise a conventional pultrusion process for forming such continuously coated extrudates. In such process, the thermoplastic material is heated to form a molten mass. A continuous fiber of the reinforcing material is pulled through this molten mass and then through a shaped orifice 220 to form a uniform coating of the thermoplastic material entirely around the fiber. In preferred processes, a plurality of the reinforcing material fibers are first collected into strands prior to the pultrusion process. Depending on the particular reinforcing material selected, these strands may include as few as two such fibers or as many as several thousand of such fibers.

Once the thermoplastic material has cooled below its heat distortion temperature, the pultruded string is cut in planes transverse to the elongation direction of the reinforcing material strands into a plurality of pellets 230, each consisting of a relatively short strand surrounded on its periphery with a layer of thermoplastic and exposed on its ends. Preferably, the length of these pellets is between about 0.60 cm and about 6.0 cm, and more preferably between about 1.3 cm and about 4.0 cm.

As will become clear as the description of this process progresses, it is not necessary that the cut be made entirely through the pultruded strings to sever same into separate and discrete pellets. Rather, it will be sufficient for this process to cut the strings to a sufficient depth to completely sever only the reinforcing material strands. Thus, the pultruded product may consist of segments

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each having a relatively short strand of the reinforcing material surrounded by a coating of the thermoplastic material, each of the segments being held together by a thin web of the thermoplastic material.

The pultrusion process may include pultruding the thermoplastic material with a plurality of separate fibers or strands of the reinforcing material, all arranged codirectionally with one another, to form a profile. The shape of these pultruded profiles will be determined by the shape of the orifice in the pultrusion die. Regardless of its shape, the pultruded profile will consist of the plurality of fibers or strands of the reinforcing material extending codirectionally in the pultrusion direction and surrounded by a substantially continuous phase of the thermoplastic material. After the thermoplastic has cooled, the profile can be cut into a multiplicity of pieces, each including relatively short strands of the reinforcing material embedded within the thermoplastic material. Again, the profile need not be entirely severed during this cutting procedure, so long as the cutting procedure completely severs all of the reinforcing strands in the profile.

The pellets or pieces 230 are then juxtaposed with one another, as at 240, so that the relatively short strands or fibers therein extend substantially codirectionally with one another in the fiber direction. The thermoplastic material in the juxtaposed pieces is then fused to form a unitary mass including a substantially continuous phase of the thermoplastic material surrounding the discontinuous fibers or strands of the reinforcing material, wherein the fibers or strands still extend codirectionally in the fiber direction. This fusing step may be performed in substantially the same manner as described above in connection with the previous process. That is, after heating the juxtaposed pieces 240 to soften the thermoplastic material (but not reinforcing material) these pieces may be subjected to a compressive load 250, applied transversely to the fiber direction, which will cause the thermoplastic material to flow together and fuse into a unitary mass 255. Although the unitary mass will include the discontinuous strands or fibers extending substantially in the

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same direction within the thermoplastic phase, these fibers or strands may be arranged in substantially end to end disposition at locations corresponding to the ends of the original individual pieces. Such end to end disposition materially reduces the physical properties of the composite. It is therefore preferable to subject the unitary mass 255 to a shearing step 260 which redistributes the discontinuous fibers or strands to side-by-side, overlapping and interweaved disposition while maintaining their substantially codirectional alignment.

In the shearing step 260, the unitary mass 255 is heated above the heat distortion temperature of the thermoplastic (but, where applicable, below the heat distortion temperature of the reinforcing material) and engaged between the confronting surfaces of a pair of opposed members while the surfaces are moved relative to one another in the fiber direction. One such shearing process may include feeding the unitary mass 255 through a nip 262 defined between a pair of opposed rollers 264 and 266 which are rotated at unequal surface velocities to form a sheet 268. As the mass passes through the nip 262, the surface in contact with the roller 266 having the greater surface velocity will be pulled relative to the surface in contact with the roller 264 having the lower surface velocity. The relative displacement of these surfaces with respect to one another will result in a redistribution of the discontinuous fibers or strands within the sheet 268, but will not affect the substantially codirectional alignment of these strands or fibers with one another.

The fusing step and shearing step may be performed in a single operation. In one such operation, the individual pellets or pieces 230 may be fed through a roll mill (not shown) having a nip defined by a pair of heated rollers rotating at unequal surface velocities. As they pass through the heated nip, the thermoplastic in each of the pieces will be heated to a flowable condition and will fuse with the thermoplastic in the adjacent pieces. At the same time, the different surface velocities of the rollers will apply a shear force to the mass to substantially redistribute the discontinuous fibers therein with respect to one another.

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The sheared, unitary sheets 268 may serve as a preform for molding composite articles to a final shape. Thus, the sheet 268 may be cut into a plurality of panels 270 which can be stacked on top of one another so that the discontinuous fibers in all of the panels extend in substantially the same direction, until a predetermined thickness is reached. The stack can then be heated to place the thermoplastic material in a flowable condition (but not the reinforcing material fibers) and compacted by applying a load transversely to the fiber direction, such as in a compression mold 280, to form an article 290 in the desired shape. This article 290 will consist of discontinuous reinforcing fibers extending codirectionally with one another and surrounded by a substantially continuous phase of thermoplastic.

The process employing the coextrusion step forms unidirectional discontinuous composites which also have physical properties which generally are greater with respect to loads applied in the fiber direction than with respect to loads applied in directions transverse to the fiber direction. The magnitude of these properties will again depend upon the particular thermoplastic and reinforcing materials from which these composites are fabricated.

In both of the processes described above, i.e., the process employing the carding step and the process employing the coextrusion step, the physical properties of the composite in the fiber direction may be further enhanced through the use of continuous reinforcing fibers which have a non-uniform cross-section in the length direction. Examples of such fibers include fibers having a diameter which modulates along the length of the fiber; twisted or spiraled fibers; fibers having a zig-zag or accordion-shaped profile; and fibers having radially protruding lobes at spaced distances along their length.

The following examples illustrate certain features of the invention as described above.

EXAMPLE 1

Extruded polypropylene fibers having a diameter of about 28 microns are collected into strands of 72 fibers each. The strands are then heated above the heat distortion temperature in an oven heated to a temperature of about 130°C and texturized by subjecting same to blasts of air. After cooling to about room temperature, the strands are cut into discrete lengths. A cardable fiberglass produced by Owens-Corning Fiberglass, Inc., 10 microns in diameter, is collected in strands of about 2,000 fibers each and cut to predetermined lengths. Cardable fiberglass is fiberglass which has been coated with a suitable surface agent or sizing which enables the strands to be unbundled into individual fibers during a carding process.

In different runs of this example, the predetermined length of the fiberglass strands are about 0.5, 1.0, 1.5, 2.0 and 2.5 inches, and the discrete length of the polypropylene strands are about 2.0 inches. By weight percentage, 30% of the cut fiberglass strands and 70% of the textured and cut polypropylene strands are loaded into a precarder in which a majority of the polypropylene and fiberglass strands are at least partially separated into individual fibers and combined in a three-dimensional fashion to yield a homogeneous mixture in which the fibers are randomly oriented.

The homogeneous mixture is then fed into a carding machine in which the fibers are pulled apart and aligned substantially codirectionally with one another in the machine direction to yield a continuous veil or web one meter wide and several fiber diameters thick at the output of the carder. The bulk density of the web ranges between about 0.2% and about 0.7% of the true density of the mixture, depending upon the strand lengths for the different runs. This web is then fed through the orifice of a die which collects the web to form a continuous sliver in which the fibers extend codirectionally parallel to the elongation direction of the sliver. The bulk density of these slivers is between about 0.004 gm/cm<sup>3</sup> and about 0.01 gm/cm<sup>3</sup>, depending upon the strand lengths and the degree of compaction imparted to the sliver during handling subsequent to the carding process.

The continuous sliver, having a diameter of about 4 cm, is then cut into lengths of about 30 cm. A plurality of these lengths are juxtaposed to form a layer 20 cm wide and 30 cm long, with the sliver lengths all extending codirectionally with one another. Thirty of such layers are stacked on top of one another to form a parallel-piped shape in which all of the sliver lengths extend in the same direction. The thus formed stack is placed between two sheets of Teflon-coated fabric and heated for about 5 minutes in an oven at a temperature of about 240°C to soften the polypropylene fibers to a flowable state. The heated stack is placed between the opposed members of a compression mold (which members are at a temperature of about 70°C) and compression molded at an applied pressure of about 200 ATM for about 30 seconds, during which time the thermoplastic fibers are fused to form a substantially continuous thermoplastic phase surrounding the discontinuous fiberglass fibers. This compacting process yields a composite 30 cm long X 20 cm wide X 3.5 mm thick.

Table 1 shows the physical properties of the composites formed in the different runs of Example 1, in both the fiber direction and in the direction transverse to the fiber direction. It can be seen that regardless of the length of the discontinuous glass fibers therein, each of the composites exhibited significantly superior strengths and toughness in the fiber direction than in the transverse direction.

Table 1

Glass Fiber Length (IN)	Flex Modulus		Flex Strength		Tensile Strength		Toughness	
	MPa		MPa		MPa		KJ/M <sup>2</sup>	
	FD	TD	FD	TD	FD	TD	FD	TD
0.5	8155	5539	193	125	93	62	58	37
1.0	8197	5315	184	118	116	71	60	36
1.5	6324	1968	154	51	86	16	50	14
2.0	6698	1747	183	58	92	19	47	12
2.5	8551	5503	191	116	84	50	59	35

FD = Fiber Direction

TD = Transverse Direction

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Although the invention herein has been described with reference to particular embodiments, it is to be understood that these embodiments are merely illustrative of the principles and applications of the present invention. It is therefore to be understood that numerous modifications may be made to the illustrative embodiments and that other arrangements may be devised without departing from the spirit and scope of the present invention as defined by the appended claims.

## CLAIMS:

1. A method of making a unidirectional discontinuous composite incorporating a thermoplastic material and an additional material comprising the steps of:

(a) forming a preform including fibers of said thermoplastic material and discontinuous fibers of said additional material intimately admixed with one another and extending generally codirectionally with one another; and

(b) fusing said thermoplastic fibers to form a thermoplastic phase surrounding said discontinuous fibers of said additional material.

2. A method as claimed in claim 1 wherein said thermoplastic fibers are discontinuous fibers

3. A method as claimed in claim 1 wherein said step of forming a preform includes the steps of mixing discontinuous fibers of said thermoplastic material and discontinuous fibers of said additional material to form a mixture and then carding said mixture so as to orient the fibers in such mixture codirectionally with one another.

4. A method as claimed in claim 3 wherein said step of forming a preform further includes the step of forming said fibers into an elongated sliver so that said codirectionally-extending fibers extend generally parallel to the direction of elongation of the sliver.

5. A method as claimed in claim 4 wherein said step of forming a preform further includes the step of juxtaposing a plurality of lengths of said sliver with one another so that said lengths of sliver extend generally parallel with one another.

6. A method as claimed in claim 1 wherein said step of fusing said thermoplastic fibers includes the step of subjecting said

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preform to heat so as to bring the thermoplastic material in said thermoplastic fibers to a flowable condition.

7. A method as claimed in claim 6 further comprising the step of compacting said preform while maintaining the thermoplastic material in a flowable condition.

8. A method as claimed in claim 7 wherein said step of compacting said preform includes the step of squeezing said preform between a pair of opposed members while said thermoplastic material is in said flowable condition.

9. A method as claimed in claim 7 wherein said step of compacting said preform includes the step of compression molding said preform.

10. A method as claimed in claim 1 wherein said additional material is selected from the group consisting of glass, carbon, ceramics, metals, non-thermoplastic polymers and thermoplastic polymers having a heat distortion temperature higher than said fibers of said thermoplastic material.

11. A method as claimed in claim 1 wherein said thermoplastic material is selected from the group consisting of polyolefins.

12. A method of making a unidirectional composite incorporating a thermoplastic material and fibers of an additional material comprising the steps of:

(a) extruding said thermoplastic material on substantially continuous fibers of said additional material to form an extrudate with said continuous fibers extending substantially in a machine direction;

(b) cutting said extrudate in a direction transverse to said machine direction to sever said continuous fibers and form an article

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having segments each including said thermoplastic material and discontinuous fibers of said additional material;

(c) fusing the thermoplastic material in said segments together to form a unitary mass including said thermoplastic material and said discontinuous fibers extending substantially in a fiber direction; and

(d) subjecting said unitary mass to shear in a direction parallel to said fiber direction while maintaining the thermoplastic material in said mass in a flowable condition to thereby redistribute said fibers in said fiber direction.

13. A method as claimed in claim 12 wherein said cutting step includes the step of severing said extrudate in a direction transverse to said machine direction to form a multiplicity of pieces each including said thermoplastic material and discontinuous fibers of said additional material, and, prior to said fusing step, said method including the step of juxtaposing said pieces with one another so that the discontinuous fibers in said pieces extend substantially codirectionally with one another in said fiber direction.

14. A method as claimed in claim 12 wherein said extruding step includes the step of coextruding said thermoplastic material with a strand including a multiplicity of said fibers.

15. A method as claimed in claim 14 wherein said coextruding step includes the step of pultruding said strand in said thermoplastic material.

16. A method as claimed in claim 12 wherein said step of subjecting said unitary mass to shear includes the step of engaging said unitary mass between confronting surfaces of a pair of opposed members while moving one of said surfaces relative to the other one of said surfaces substantially in said fiber direction.

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17. A method as claimed in claim 16 wherein said step of subjecting said unitary mass to shear includes the step of passing said mass through a nip between a pair of rollers while rotating said rollers.

18. A method as claimed in claim 12 wherein said additional material is selected from the group consisting of glass, ceramics, metals, non-thermoplastic polymers and thermoplastic polymers having a heat distortion temperature higher than said fibers of said thermoplastic material.

19. A method as claimed in claim 12 wherein said thermoplastic material is selected from the group consisting of polyolefins.

20. A method as claimed in claim 12 wherein said fusing step is performed so that at least some of said discontinuous fibers in said mass are disposed in substantially end-to-end relation with other fibers in said mass, and wherein said step of subjecting the mass to shear redistributes at least some of said fibers from said end-to-end relation to overlapping relation with one another.

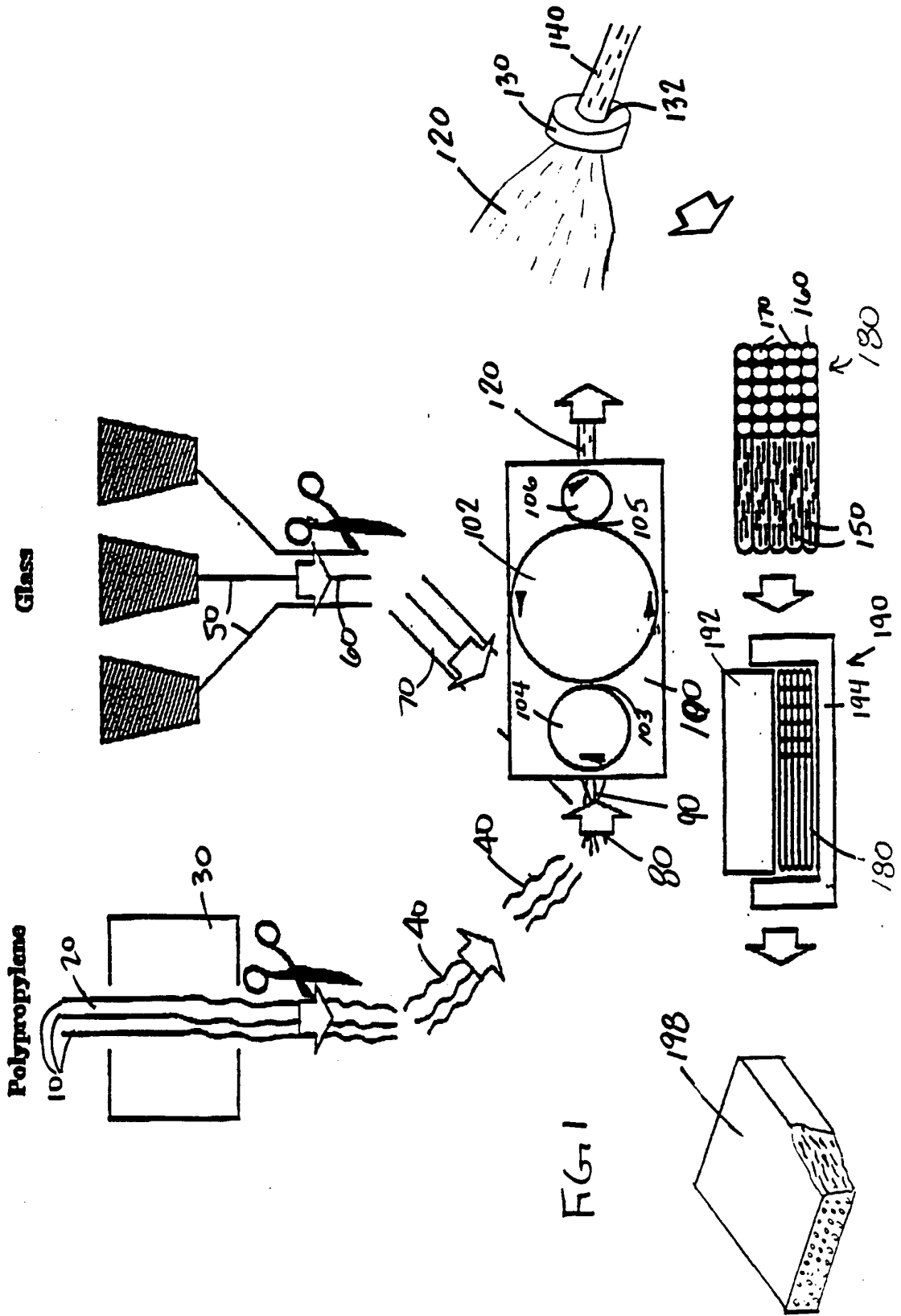


FIG 1

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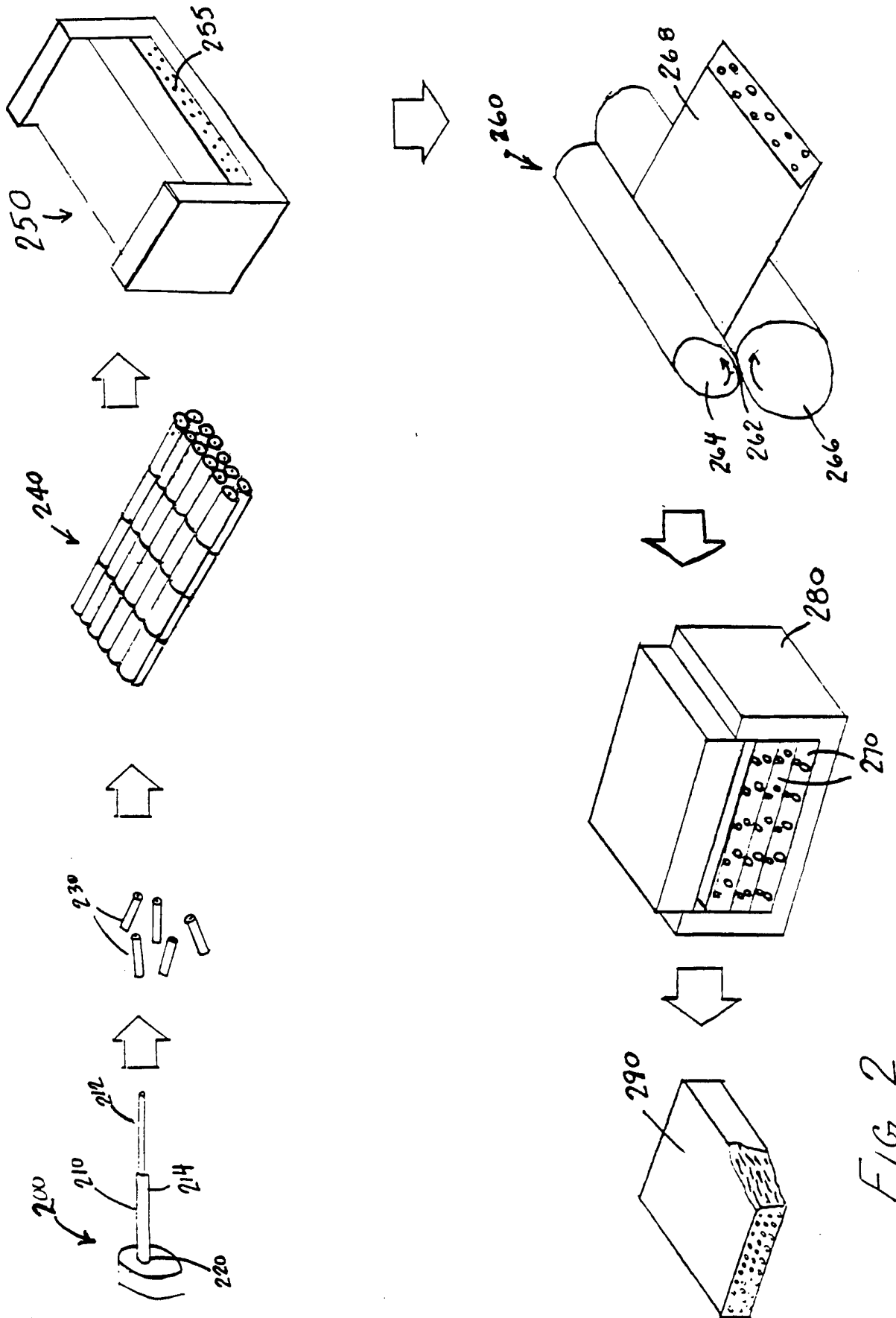


FIG 2