

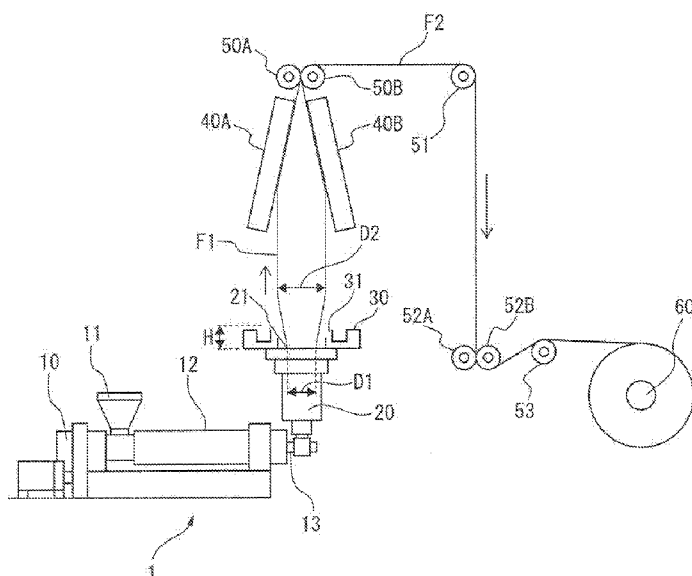


- (51) International Patent Classification:  
*B29C 55/28* (2006.01) *B29C 49/04* (2006.01)
- (21) International Application Number:  
PCT/US2016/017288
- (22) International Filing Date:  
10 February 2016 (10.02.2016)
- (25) Filing Language: English
- (26) Publication Language: English
- (71) Applicants: **THE YOKOHAMA RUBBER CO., LTD.** [JP/JP]; 36-11, Shimbashi 5-chome, Minato-ku, Tokyo, 1058685 (JP). **EXXONMOBIL CHEMICAL PATENTS INC.** [US/US]; 4500 Bayway Drive, Baytown, TX 77520 (US).
- (72) Inventors: **HARA, Yuichi**; c/o The Yokohama Rubber Co., Ltd., Hiratsuka Factory, 2-1, Oiwake, Hiratsuka-shi, Kanagawa, 254-8601 (JP). **TOMOI, Shusaku**; c/o The Yokohama Rubber Co., Ltd., Hiratsuka Factory, 2-1, Oiwake, Hiratsuka-shi, Kanagawa, 254-8601 (JP). **SATO, Shun**; c/o The Yokohama Rubber Co., Ltd., Hiratsuka Factory, 2-1, Oiwake, Hiratsuka-shi, Kanagawa, 254-8601 (JP). **NADELLA, Hari, P.**; 3561 Black Cherry Point SW, Gainesville, GA 30504 (US).
- (74) Agent: **AMERNICK, Burton, A.**; Novak Druce Connolly Bove + Quigg LLP, 1875 Eye Street, NW, Suite 1100, Washington, DC 20006 (US).
- (81) Designated States (*unless otherwise indicated, for every kind of national protection available*): AE, AG, AL, AM, AO, AT, AU, AZ, BA, BB, BG, BH, BN, BR, BW, BY, BZ, CA, CH, CL, CN, CO, CR, CU, CZ, DE, DK, DM, DO, DZ, EC, EE, EG, ES, FI, GB, GD, GE, GH, GM, GT, HN, HR, HU, ID, IL, IN, IR, IS, JP, KE, KG, KN, KP, KR, KZ, LA, LC, LK, LR, LS, LU, LY, MA, MD, ME, MG, MK, MN, MW, MX, MY, MZ, NA, NG, NI, NO, NZ, OM, PA, PE, PG, PH, PL, PT, QA, RO, RS, RU, RW, SA, SC, SD, SE, SG, SK, SL, SM, ST, SV, SY, TH, TJ, TM, TN, TR, TT, TZ, UA, UG, US, UZ, VC, VN, ZA, ZM, ZW.
- (84) Designated States (*unless otherwise indicated, for every kind of regional protection available*): ARIPO (BW, GH, GM, KE, LR, LS, MW, MZ, NA, RW, SD, SL, ST, SZ, TZ, UG, ZM, ZW), Eurasian (AM, AZ, BY, KG, KZ, RU, TJ, TM), European (AL, AT, BE, BG, CH, CY, CZ, DE, DK, EE, ES, FI, FR, GB, GR, HR, HU, IE, IS, IT, LT, LU, LV, MC, MK, MT, NL, NO, PL, PT, RO, RS, SE, SI, SK, SM, TR), OAPI (BF, BJ, CF, CG, CI, CM, GA, GN, GQ, GW, KM, ML, MR, NE, SN, TD, TG).

[Continued on next page]

(54) Title: INFLATION MOLDING EQUIPMENT FOR PRODUCING CYLINDRICAL FILM

FIG. 1



(57) Abstract: Inflation molding equipment and a process for producing a cylindrical film are disclosed. The inflation molding equipment comprises an extruder for melt extruding a stock material, a single-layer or multi-layer cylindrical die attached to a discharge port of the extruder, and an air ring device for cooling a cylindrical film extruded from the cylindrical die, wherein the cylindrical die has an annular discharge port having an outer diameter of D1, the air ring device is attached to the cylindrical die, and the air ring device has at least one cooling gas outlet port, the cooling gas outlet port having a cooling gas channel defined by an outer circumferential wall having a height of H and an inner circumferential wall having a height; lower than the outer circumferential wall, and D1 and H satisfy a relationship of  $Q \cdot 5xD1 \leq H \leq 1.5xD1$ .

**WO 2017/138930 A1**



---

**Published:**

— *with international search report (Art. 21(3))*

- 1 -

## DESCRIPTION

Title of Invention: Inflation Molding Equipment for  
Producing Cylindrical Film

5

Technical Field

[0001] The present invention relates to an inflation  
molding equipment and a process for producing a  
cylindrical film. More specifically, the present  
10 invention relates to an inflation molding (blown film  
extrusion molding) equipment capable of producing a  
cylindrical film having excellent quality with a high  
dimensional precision, a process for producing a  
cylindrical film using the inflation molding equipment,  
15 and a process for producing a pneumatic tire using the  
cylindrical film.

Background Art

[0002] It is known to extrude a melt of a  
thermoplastic resin composition or a thermoplastic  
20 elastomer composition comprising as a continuous phase  
(sea phase) a thermoplastic resin and as a disperse phase  
(island phase) an elastomer dispersed in the  
thermoplastic resin by an extrusion molding equipment  
into a molded article in the form of film, etc. (see PLT  
25 1). When a cylindrical film is produced from such a  
composition by the inflation molding method, in the  
process in which a cylindrical film (hereinafter referred  
to also as a "bubble") in a softened state melt-extruded  
from a cylindrical die is drawn in the vertical direction  
30 to thereby expanding the bubble while cooling it, the  
bubble is stretched in the vertical direction (machine  
direction or conveyance direction) and transverse  
direction (circumferential direction) perpendicular to

- 2 -

the vertical direction. When a cylindrical film is produced from a thermoplastic elastomer composition, if the elastomer particles present as a disperse phase in the thermoplastic elastomer composition are drawn and stretched in the vertical and transverse directions of the bubble in the process of expanding the bubble melt-extruded from a cylindrical die, and accordingly, when the drawing force applied to the cylindrical film is released after inflation molding, the cylindrical film easily shrinks, and consequently the resulting cylindrical film has low dimensional precision, i.e., low precision to a target dimension value. Further, the resulting film has a problem that the film has wrinkles or waviness due to shrinkage. Therefore, when a cooling gas is blown from an air ring device to the bubble to cool down the bubble, it is desirable to expand the bubble to a target maximum diameter while gradually cooling the bubble. In order to improve productivity and yield, it is desirable to expand the bubble to a target maximum diameter while gradually cooling the bubble without impairing the stability of the bubble.

## Citations List

## Patent Literature

[0003] PLT 1: Japanese Unexamined Patent Publication (JP-A) No. 2015-143317

## Summary of Invention

## Technical Problem

[0004] In view of the above conventional problems, the present invention is directed to provide an inflation molding equipment capable of improving the stability of a bubble and capable of reducing shrinkage of the cylindrical film, and as a result capable of producing a cylindrical film with a high dimensional precision.

- 3 -

[0005] The inventors have carried out experiments to achieve the above object, and as a result found that, in an inflation molding equipment for producing a cylindrical film, when an outer diameter D1 of an annular discharge port of a cylindrical die and a height H of an outer circumferential wall of a cooling gas outlet port of an air ring device for cooling a bubble extruded from the annular discharge port satisfy a specific relationship, it is possible to improve the stability of the bubble and to reduce shrinkage of the cylindrical film, and as a result it is possible to produce a cylindrical film having a high dimensional precision, and consequently completed the present invention.

Solution to Problem

[0006] The present invention includes the following [Embodiment 1] to [Embodiment 7].

[Embodiment 1] An inflation molding equipment comprising:

an extruder for melt extruding a stock material,  
a single-layer or multi-layer cylindrical die attached to a discharge port of the extruder, and  
an air ring device for cooling a cylindrical film extruded from the cylindrical die, wherein  
the cylindrical die has an annular discharge port having an outer diameter of D1,  
the air ring device is attached on the cylindrical die, and  
the air ring device has at least one cooling gas outlet port, the cooling gas outlet port having a cooling gas channel defined by an outer circumferential wall having a height of H and an inner circumferential wall having a height lower than the outer circumferential wall, and

- 4 -

D1 and H satisfy a relationship of the following formula (1):

$$0.5 \times D1 \leq H \leq 1.5 \times D1 \quad (1).$$

[0007] [Embodiment 2] The inflation molding equipment according to Embodiment 1, wherein the stock material is a thermoplastic elastomer composition having an island-in-sea structure comprised of a continuous phase comprising a thermoplastic resin and a disperse phase comprising an elastomer.

10 [0008] [Embodiment 3] The inflation molding equipment according to Embodiment 1, wherein D1 and H satisfy a relationship of the following formula (2):

$$0.8 \times D1 \leq H \leq 1.2 \times D1 \quad (2).$$

15 [0009] [Embodiment 4] The inflation molding equipment according to any one of Embodiments 1 to 3 wherein a ratio of an effective length L of a screw to a screw diameter D of the extruder, L/D, is 32 or more.

[0010] [Embodiment 5] The inflation molding equipment according to any one of Embodiments 1 to 4, wherein the air ring device does not have a rectifying cylinder spaced apart from the outer circumferential wall of the cooling gas outlet port at the outer circumferential side of the outer circumferential wall.

20 [0011] [Embodiment 6] A process for producing a cylindrical film, comprising:

providing the inflation molding equipment according to any one of Embodiments 1 to 5;

30 melt extruding a molten thermoplastic elastomer composition from the extruder to the cylindrical die, the molten thermoplastic elastomer composition comprising a thermoplastic resin selected from the group consisting of a polyamide-based resin, a polyvinyl-based resin, a polyester-based resin, and a blend of two or more of

- 5 -

these resins as a continuous phase, and a modified elastomer as a disperse phase; and

extruding the molten thermoplastic elastomer composition as a cylindrical film from the cylindrical die.

[0012] [Embodiment 7] A process for producing a pneumatic tire comprising:

producing a cylindrical film by the process according to Embodiment 6;

cutting the resulting film in accordance with a size of the pneumatic tire to form an inner liner;

forming a green tire comprising the inner liner; and

vulcanizing the green tire to form a pneumatic tire.

[0013] Advantageous Effects of Invention

According to the inflation molding equipment of the present invention, it is possible to improve the stability of a bubble and to reduce shrinkage of the cylindrical film, and as a result, it is possible to produce a cylindrical film with a high dimensional precision. The cylindrical film produced using the inflation molding equipment of the present invention has excellent features of a high dimensional precision and few wrinkles or waviness on the surface thereof. Further, since the inflation molding equipment of the present invention can produce a cylindrical film having a high dimensional precision without arranging a rectifying cylinder spaced apart from the outer circumferential wall of the cooling gas outlet port of the air ring device as described in Embodiment 1 at the outer circumferential side of the outer circumferential wall, and therefore it is possible to eliminate a preliminary adjustment of the inflation molding equipment accompanying use of a conventional rectifying cylinder and to reduce the costs

- 6 -

or work involved in the rectifying cylinder.

#### Brief Description of Drawings

[0014] [FIG. 1] FIG. 1 is a schematic view showing one embodiment of a process for producing a cylindrical film using an inflation molding equipment according to the present invention.

[FIG. 2] FIG. 2 is a schematic view showing another embodiment of a process for producing a cylindrical film using an inflation molding equipment according to the present invention.

[FIG. 3] FIG. 3 is a schematic view showing a mechanism of cooling a cylindrical film by an air ring device when producing a cylindrical film using an inflation molding equipment according to the present invention.

#### Description of Embodiments

[0015] Embodiments of the present invention will be explained below in detail along referring to the attached drawings. FIG. 1 shows one embodiment of an inflation molding equipment according to the present invention. This inflation molding equipment 1 comprises extruder 10 comprising stock material feeder 11, cylinder 12 and discharge port 13, and cylindrical die 20 connected to discharge port 13 of extruder 10. A melt-extrudable stock material is introduced into cylinder 12 set to a melt extrusion temperature from stock material feeder 11 of extruder 10, and the stock material is allowed to melt in cylinder 12 while extruding it by a rotating screw (not shown) to the discharge port side, and then the resulting molten stock material is extruded from discharge port 13 to cylindrical die 20. Cylindrical die 20 has an annular discharge port 21 having an outer diameter D1. The molten stock material introduced into cylindrical die 20 is extruded from annular discharge port 21 as a cylindrical

- 7 -

film (bubble) F1 in a softened state, and cylindrical film F1 gradually expands until the diameter thereof reaches the maximum diameter D2 while being cooled along with being conveyed upward. The term "the diameter of the cylindrical film" refers herein to the outer diameter of the cylindrical film. The target value of D2 is determined in accordance with the desired size of the cylindrical film. Hereinafter, the ratio of D2 to D1, D2/D1, will be referred to as a "blow ratio". In FIG. 1, a pair of stabilizing plates 40A, 40B facing each other are provided above air ring device 30. Cylindrical film F1 is cooled along with being conveyed upward and is deformed into flat while being conveyed by the pair of stabilizing plates 40A, 40B facing each other. In place of the pair of stabilizing plates 40A, 40B, for example, a plurality of guide rollers arranged in parallel with each other in a direction perpendicular to the conveyance direction of bubble B may be used. Above stabilizing plates 40A, 40B, a pair of pinch rolls 50A, 50B are arranged for folding cylindrical film F1 deformed into flat by stabilizing plates 40A, 40B, into a sheet. Cylindrical film (lay-flat film) F2 which is obtained by folding the cylindrical film by the pair of pinch rolls 50A, 50B is wound up by windup roll 60 through guide rolls 51, 52A, 52B, 53. Before cylindrical film F2 is wound up by the windup roll or after it is wound up, one end of cylindrical film F2 may be cut open to obtain a wide film, or both ends of cylindrical film F2 may be slit to obtain two films, as necessary.

[0016] FIG. 2 shows another embodiment of an inflation molding equipment according to the present invention. This inflation molding equipment comprises a first extruder 10A for extruding the molten first stock

- 8 -

material and a second extruder 10B for extruding the molten second stock material. The first extruder 10A and the second extruder 10B respectively comprise stock material feeder 11A, 11B, cylinder 12A, 12B, and discharge port 13A, 13B. A multilayer cylindrical spiral die is attached to each of discharge port 13A of the first extruder 10A and discharge port 13B of the second extruder 10B. The stock material melt extruded from the first extruder and the stock material melt extruded from the second extruder may be the same or different. A multilayer film can be produced using an extrusion molding equipment having a configuration as shown in FIG. 2.

[0017] FIG. 3 is a view schematically showing a mechanism for cooling a cylindrical film by the air ring device when using an inflation molding equipment according to the present invention to produce a cylindrical film. In FIG. 3, one embodiment of the cylindrical die and the air ring device is shown by a schematic vertical cross-sectional view. The air ring device schematically shown in FIG. 3 is a "dual lip" type air ring device and has two annular cooling gas outlet ports. Air ring device 30 is mounted on cylindrical die 20. Air ring device 30 has annular plenum chamber 31. A cooling gas is fed from an external supply source (not shown) to annular plenum chamber 31. The cooling gas fed to annular plenum chamber 31 passes through annular channel 32 to lower annular outlet port 33 and upper annular outlet port 34. The cooling gas is blown from lower annular outlet port 33 and upper annular outlet port 34 to the cylindrical film. Upper annular outlet port 34 has cooling gas channel 35 defined by outer circumferential wall 34A and inner circumferential wall

- 9 -

34B having a height H lower than that of the outer circumferential wall. Lower annular outlet port 33 and upper annular outlet port 34 are arranged concentrically and have a common center axis. The direction of this  
5 common center axis coincides with the direction of the center axis of the cylindrical die or the direction at which the bubble extruded from the cylindrical die is drawn. When the air ring device has two or more annular cooling gas outlet ports, the outer circumferential walls  
10 of the cooling gas outlet ports are preferably higher in height from the common center axis toward the outer circumferential sides. That is, it is preferable that a plurality of cooling gas outlet ports are arranged concentrically so that the line connecting the topmost  
15 parts of the outer circumferential walls of the plurality of cooling gas outlet ports forms a U-shape or recessed shape with respect to the direction at which the molten stock material extruded from the cylindrical die is drawn. The "height H of the outer circumferential wall"  
20 refers herein to the distance from the top surface of the cylindrical die at which the air ring device is mounted to the topmost part of the outer circumferential wall. When the air ring device has a plurality of cooling gas outlet ports, the "height H of the outer circumferential  
25 wall" means the highest height of the outer circumferential wall at the outermost side. The topmost part of the outer circumferential wall having the height H is preferably arranged at a position spaced apart from the inside wall of the outer lip of the cylindrical die  
30 by a distance of from 12.5 to 88 mm in a direction perpendicular to the direction of the center axis of the cylindrical die. The outer circumferential wall of the cooling gas outlet port may be configured to enable

- 10 -

adjustment of the height H. For example, the outer circumferential wall of a cooling gas outlet port may be comprised of an annular base part and an annular component part connected to the annular base part. For example, when the annular base part and the annular component part have mutually engaging threaded parts, the height H can be adjusted by rotating the annular component part while engaging the annular base part with the annular component part. Air ring device 30 has a gas inlet port (not shown) for taking in a cooling gas from an external supply source to the inside of the air ring device. The cooling gas can be selected from air and an inert gas (for example, nitrogen and argon), etc. Although the mechanism for cooling the cylindrical film by the air ring device is schematically illustrated along referring to FIG. 3, the configuration of the air ring device is not limited to the embodiment shown in FIG. 3. In the inflation molding equipment of the present invention, it is possible to produce a cylindrical film having a high dimensional precision without arranging a rectifying cylinder at the outer circumferential side of the outer circumferential wall of the cooling gas outlet port of the air ring device while spacing apart from the outer circumferential wall, as described above. When a rectifying cylinder is arranged spaced apart from the outer circumferential wall at the outer circumferential side of the outer circumferential wall of the cooling gas outlet port, the bubble is stabilized due to the venturi effect, but resulting in an easily shrinkable cylindrical film. For this reason, in the inflation molding equipment of the present invention, the air ring device preferably does not have a rectifying cylinder spaced apart from the outer circumferential wall of the cooling gas outlet port

- 11 -

at the outer circumferential side of the outer circumferential wall.

[0018] In the inflation molding equipment of the present invention, the outer diameter D1 of the annular discharge port of the cylindrical die and the height H of the outer circumferential wall of the air ring device satisfy the relationship of the following formula (1):

$$0.5 \times D1 \leq H \leq 1.5 \times D1 \quad (1).$$

Preferably, D1 and H satisfy the relationship of the following formula (2):

$$0.8 \times D1 \leq H \leq 1.2 \times D1 \quad (2).$$

By satisfying the relationship of the above formula (1), the stability of the bubble can be improved while the shrinkage of the cylindrical film can be reduced, and as a result a cylindrical film can be produced having high dimensional precision. When the relationship of the above formula (1) is satisfied, it is believed that the gradual cooling of the cylindrical film (bubble) in a softened state which is melt-extruded from the cylindrical die can be carried out effectively until the film expands to the diameter D2. If H is less than  $0.5 \times D1$ , it is difficult to form a bubble steadily, and therefore, it is difficult to produce a cylindrical film having high precision. If H is more than  $1.5 \times D1$ , the dimensional precision is decreased and wrinkling or waviness occurs in the film. The value of D1 is determined in accordance with the desired size of the cylindrical film. The size of D1 is not particularly limited, but in the case that the cylindrical film produced by the inflation molding equipment of the present invention is used, for example, as an inner liner for the pneumatic tire of a passenger car, D1 is typically from 400 mm to 800 mm. In the inflation molding equipment of the present invention, so

- 12 -

long as D1 and H satisfy the relationship of the above formula (1), the values of D1 and H are not particularly limited, but for example in the case of an 8 inch die, D1 is 203 mm, and in that case, H is from 102 mm to 304 mm, preferably from 142 mm to 264 mm, and more preferably from 162 mm to 244 mm.

[0019] The film produced using the inflation molding equipment of the present invention preferably has a thickness of 0.01 to 0.2 mm. The film produced using the inflation molding equipment of the present invention can achieve a sufficient air pressure retention rate sufficient, when it is used, for example, as an inner liner of a pneumatic tire at such a thickness.

[0020] A thermoplastic resin composition or a thermoplastic elastomer composition comprising a thermoplastic resin as a continuous phase (sea phase) and an elastomer as a disperse phase (island phase), in which the elastomer is dispersed in the thermoplastic resin, can be used as the stock material for producing a film by the inflation molding equipment of the present invention. Examples of the thermoplastic resin which can form the thermoplastic resin composition or thermoplastic elastomer composition include polyamide-based resins, polyvinyl-based resins, polyester-based resins, polynitrile-based resins, polymethacrylate-based resins, cellulosic resins, fluororesins, imide-based resins, polystyrenic resins, polyolefinic resins, etc. The thermoplastic resin composition or thermoplastic elastomer composition may include at least one thermoplastic resin. The film produced from the thermoplastic elastomer composition is lighter in weight compared with an inner liner based on conventional butyl rubber and, further, has an air pressure retention rate

- 13 -

which is equal to or higher than that of an inner liner based on conventional butyl rubber, and therefore it is preferable to produce a pneumatic tire inner liner from a thermoplastic elastomer composition. When a thermoplastic elastomer composition is used as the stock material, the thermoplastic elastomer composition preferably comprises a combination of a thermoplastic resin having high barrier properties and a modified elastomer having a high affinity to the thermoplastic resin.

5 [0021] Examples of polyamide-based resins include, Nylon 6 (N6), Nylon 66 (N66), Nylon 46 (N46), Nylon 11 (N11), Nylon 12 (N12), Nylon 69 (N69), Nylon 610 (N610), Nylon 612 (N612), Nylon 6/66 copolymer (N6/66), Nylon 6/610 copolymer (N6/610), Nylon 6/66/610 copolymer (N6/66/610); semi-aromatic and all-aromatic nylons such as Nylon MXD6 (MXD6), Nylon 6T, Nylon 9T, Nylon 6/6T copolymer; Nylon 66/PP copolymer, Nylon 66/PPS copolymer, a copolymer of a polyamide and polyether (for example, a copolymer of at least one type of the above Nylon and a polyether), and *N*-alkoxyalkylates thereof, for example methoxymethylate of Nylon 6, methoxymethylate of Nylon 6/610, methoxymethylate of Nylon 612, etc. Examples of polyvinyl-based resins include ethylene-vinyl acetate copolymer (EVA), poly(vinyl alcohol) (PVA), vinyl alcohol-ethylene copolymer (EVOH), poly(vinylidene chloride) (PVDC), poly(vinyl chloride) (PVC), vinyl chloride/vinylidene chloride copolymer, vinylidene chloride/methylacrylate copolymer, vinylidene chloride/acrylonitrile copolymer, etc. Examples of polyester-based resins include polybutylene terephthalate (PBT), polyethylene terephthalate (PET), polyethylene isophthalate (PEI), PET/PEI copolymer, polyarylate (PAR), polybutylene naphthalate (PBN), liquid crystal

- 14 -

polyesters, polyoxyalkylenediimidic acid/polybutylate terephthalate copolymers, or other aromatic polyesters, a copolymer of a polyester and polyether, a copolymer of a polyester and an aliphatic polycarbonate, etc. Examples of polynitrile-based resins include polyacrylonitrile (PAN), polymethacrylonitrile, acrylonitrile/styrene copolymer (AS), methacrylonitrile/styrene copolymer, methacrylonitrile/styrene/butadiene copolymer, etc. Examples of polymethacrylate-based resins include poly(methyl methacrylate) (PMMA), poly(ethyl methacrylate), etc. Examples of cellulosic resins include cellulose acetate, cellulose acetate butyrate, etc. Examples of fluororesins include poly(vinylidene fluoride) (PVDF), poly(vinyl fluoride) (PVF), polychlorofluoroethylene (PCTFE), tetrafluoroethylene/ethylene copolymer (ETFE), etc. Examples of imide-based resins include aromatic polyimides (PI), etc. Examples of polystyrenic resins include polystyrene (PS), etc. Examples of polyolefinic resins include polyethylene (PE), polypropylene (PP), etc. Among these thermoplastic resins, Nylon 6, Nylon 66, Nylon 46, Nylon 11, Nylon 12, Nylon 69, Nylon 610, Nylon 612, Nylon 6/66, Nylon 6/66/12, and Nylon 6/66/610 and semi-aromatic and all-aromatic nylons such as Nylon MXD6, Nylon 6T, Nylon 9T, and Nylon 6/6T are preferred, in view of gas barrier properties.

[0022] Common ingredients for common thermoplastic resin compositions, such as fillers, reinforcing agents, processing aids, stabilizers, antioxidants, etc., may optionally be added at a common amount to the thermoplastic resin which constitutes the thermoplastic resin composition or thermoplastic elastomer composition which is extruded using the inflation molding equipment

- 15 -

according to the present invention. In view of the gas barrier properties and heat resistance of the film obtained by extrusion molding of the thermoplastic resin composition or thermoplastic elastomer composition, it is preferred to not add a plasticizer to the thermoplastic resin composition or thermoplastic elastomer composition. A film having a reduced shrinkage after extrusion molding can be produced even if a plasticizer such as, for example, higher alcohol-based plasticizers, aromatic sulfoneamide-based plasticizers, phenolic plasticizers, etc., conventionally used in the extrusion molding of a thermoplastic resin composition or thermoplastic elastomer composition, is not incorporated into the thermoplastic resin composition or thermoplastic elastomer composition, and therefore it is possible to prevent the decrease in the gas barrier properties and heat resistance which may be caused when a plasticizer is added to a thermoplastic resin composition or thermoplastic elastomer composition.

[0023] The thermoplastic elastomer composition comprises at least one rubber dispersed in at least one thermoplastic resin, wherein the at least one thermoplastic resin constitutes a matrix phase (or a continuous phase) and the rubber constitutes a disperse phase (or a discontinuous phase). The rubber is dispersed in the form of particles in the thermoplastic resin. Examples of the rubber which can constitute the thermoplastic elastomer composition include diene-based rubbers, olefin-based rubbers, and their hydrogenates, epoxy-modified products, carboxylic acid-modified products, halogenates, etc., for example, natural rubber, polyisoprene, polybutadiene, styrene-butadiene copolymer, butadiene rubber, acrylonitrile-butadiene copolymer,

- 16 -

epoxydized natural rubber, hydrogenated NBR, hydrogenated SBR, ethylene-propylene rubber, ethylene-propylene-diene rubber, ethylene-glycidyl methacrylate copolymer, ethylene-alkylacrylate copolymer, ethylene-methacrylate copolymer, polybutene, polyisobutylene, maleic anhydride-modified ethylene-propylene rubber, maleic anhydride-modified ethylene- $\alpha$ -olefin copolymer, succinic anhydride-modified polyisobutylene, maleic anhydride-modified ethylene-ethylacrylate copolymer, butyl rubber, a copolymer of isobutylene and aromatic vinyl or diene-based monomer (for example, isobutylene-*p*-alkylstyrene copolymer, styrene-isobutylene-styrene terpolymer and hydrogenates thereof, acid anhydride-modified styrene-isobutylene-styrene block copolymer, etc.), brominated butyl rubber (Br-IIR), chlorinated butyl rubber (Cl-IIR), brominated isobutylene-*p*-methylstyrene copolymer (Br-IPMS), halogenated isobutylene-isoprene copolymer rubber, etc. Among these rubbers, brominated isobutylene-*p*-methylstyrene copolymer, maleic anhydride-modified ethylene- $\alpha$ -olefin copolymer, and succinic anhydride-modified polyisobutylene are preferred in view of gas barrier properties, durability and processability.

[0024] Examples of the combination of the thermoplastic resin and rubber, capable of forming the thermoplastic elastomer composition which can be used in the present invention include a combination of (a) a polyamide-based resin, (b) an ethylene-vinyl alcohol copolymer, or (c) a blend of a polyamide-based resin and ethylene-vinyl alcohol copolymer, as a thermoplastic resin, and (i) butyl rubber, (ii) a maleic anhydride-modified ethylene- $\alpha$ -olefin copolymer, or (iii) a blend of brominated isobutylene-*p*-methylstyrene copolymer rubber

- 17 -

and maleic anhydride-modified ethylene- $\alpha$ -olefin copolymer, as a rubber, i.e., a combination of a thermoplastic resin selected from (a) to (c) and a rubber selected from (i) to (iii). The combination of butyl  
5 rubber having excellent gas barrier properties and a polyamide-based resin is preferred. Among these, a combination of a brominated isobutylene-*p*-methylstyrene copolymer rubber, which is a modified butyl rubber, and one or more polyamide-based resins (for example Nylon 6,  
10 Nylon 6/66, Nylon 612, etc.) is specifically preferred in view of achieving both fatigue resistance and gas barrier properties.

[0025] The rubber particles included in the thermoplastic elastomer composition may contain carbon  
15 black, silica, or other reinforcing agents (fillers), cross-linking agent, antioxidant, processing aid, or other compounding agents that are commonly blended into a rubber composition to the extent that the effects of the present invention are not impaired.

20 [0026] Before the thermoplastic elastomer composition is introduced into extruder 10, the thermoplastic elastomer composition can be prepared in advance by melt-kneading at least one thermoplastic resin, at least one rubber, and optionally additives by, for example, a  
25 single-screw or twin-screw kneading extruder, to disperse the rubber particles as a disperse phase in the thermoplastic resin which forms a matrix phase. The weight ratio of the thermoplastic resin to the rubber is preferably from 10/90 to 90/10, and more preferably from  
30 15/85 to 90/10, but is not limited thereto. In order to fix the dispersed state of the rubber in the thermoplastic resin, the rubber is preferably dynamically cross-linked while melt-kneading the thermoplastic resin

- 18 -

and rubber. The term "dynamic cross-linking" refers herein to cross-linking simultaneous with melt-kneading. The melt-kneading temperature may be equal to or higher than the melting point of the thermoplastic resin, and is preferably a temperature which is higher than the melting point of the thermoplastic resin by 20°C, for example, is from 200 to 250°C. The total time of the kneading operation is not particularly limited, but is usually from 1 minute to 10 minutes. The thermoplastic elastomer composition which is obtained after melt-kneading the thermoplastic resin and the rubber can be extruded into, for example, a strand form, and then pelletized with a resin pelletizer.

[0027] The cross-linking agent can be suitably selected depending on the type of rubber and is not particularly limited. Examples of the cross-linking agent include zinc oxide, stearic acid, zinc stearate, magnesium oxide, *m*-phenylene bismaleimide, alkylphenol resin, and halogenates thereof, a polymerized 2,2,4-trimethyl-1,2-dihydroquinoline, etc. Among these cross-linking agents, zinc oxide, stearic acid, and calcium stearate are preferable. The amount of the cross-linking agent is preferably 0.1 to 12 parts by weight, and more preferably 1 to 9 parts by weight, with respect to 100 parts by weight of the rubber.

[0028] The thermoplastic elastomer composition which can be extrusion molded using the inflation molding equipment of the present invention can be formed in-situ by introducing the thermoplastic resin, rubber, and optionally additives from stock material feeder 11 of extruder 10 which is a single-screw or twin-screw kneading extruder, to the inside of cylinder 12, and melt-kneading them by means of rotating single or double

- 19 -

screws. In this case, the resulting thermoplastic elastomer composition can be extruded as a cylindrical film without being pelletized, from the cylindrical die attached to the discharge port of the extruder.

5     [0029]     The film produced using the inflation molding equipment of the present invention is useful as an inner liner of a pneumatic tire due to excellent gas barrier properties thereof. Any conventional process may be used as the process for producing a pneumatic tire. For  
10     example, when the film produced using the inflation molding equipment of the present invention is used as an inner liner in the production of a pneumatic tire, a pneumatic tire can be produced by laminating the film produced using the inflation molding equipment of the  
15     present invention onto a tire molding drum in cylindrical form; sequentially laminating thereon tire members such as a carcass layer, a belt layer, a tread layer, etc., to form a green tire; removing the resulting green tire from the tire molding drum, and subsequently vulcanizing the  
20     green tire according to a conventional method. The film produced using the inflation molding equipment of the present invention has a reduced shrinkage as described above, and accordingly has fewer wrinkles and is excellent in dimensional stability. Therefore, the film  
25     produced using the inflation molding equipment of the present invention can suppress defects due to the wrinkles and delamination of the film, thereby reducing the failure rate of the tire after vulcanization.

#### Examples

30     [0030]     The present invention will be further explained with reference to the following examples, and it should be understood that the scope of the present invention is not limited by these examples.

- 20 -

[0031] [Preparation of Thermoplastic Elastomer Composition]

Among the stock materials shown in Table 1 below, a brominated isobutylene *p*-methylstyrene copolymer (Br-IPMS) was processed into pellets in advance by a rubber pelletizer (manufactured by Moriyama Works). The resulting pellets of Br-IPMS and the other elastomers, thermoplastic resins, and additives (that is, plasticizer, antioxidant, and cross-linking agent) were charged into a twin-screw extruder (manufactured by Japan Steel Works) at the compounding ratio (unit: "phr", parts by weight with respect to 100 parts by weight of total elastomers) shown in Table 1 and were kneaded at 250°C for 3 minutes. The resulting kneaded mass was continuously extruded into a strand form from the extruder, cooled with water, and subsequently cut by a cutter to obtain a thermoplastic elastomer composition in the form of pellets.

[0032] Table 1

|                     |                                                                | Amount (phr) |
|---------------------|----------------------------------------------------------------|--------------|
| Elastomer           | Brominated isobutylene <i>p</i> -methylstyrene Copolymer       | 70           |
|                     | Maleic anhydride-modified ethylene- $\alpha$ -olefin copolymer | 20           |
|                     | Succinic anhydride-modified Polyisobutylene                    | 10           |
| Thermoplastic resin | Nylon 6                                                        | 40           |
|                     | Nylon 6/66                                                     | 6            |
|                     | Nylon 6/12                                                     | 12           |
|                     | Ethylene-vinyl alcohol copolymer                               | 3            |
| Ingredients         | Antioxidant ( <i>p</i> -phenylenediamine)                      | 1.0          |
|                     | Zinc oxide                                                     | 0.7          |
|                     | Stearic acid                                                   | 10           |
|                     | Calcium stearate                                               | 1.0          |

20

- 21 -

**[0033]** Footnote of Table 1:

Brominated isobutylene-*p*-methylstyrene copolymer (Br-IPMS): Exxpro® MDX89-4 from ExxonMobil Chemical Company)  
Maleic anhydride-modified ethylene- $\alpha$ -olefin copolymer:  
5 Tafmer® MH7010 from Mitsui Chemicals  
Succinic anhydride-modified polyisobutylene: DOVERMULSE H1000 fromDover Chemical Corporation  
Nylon 6: UBE Nylon® 1013FB from Ube Industries, Ltd.  
Nylon 6/66 copolymer: UBE Nylon® 5023B from Ube  
10 Industries, Ltd.  
Nylon 6/12 copolymer: UBE Nylon® 7024B from Ube Industries, Ltd.  
Ethylene-vinyl alcohol copolymer: H4815B from Nippon Synthetic Chemical Industry  
15 Antioxidant: *N*-(1,3-dimethylbutyl)-*N'*-phenyl-*p*-phenylenediamine (Santoflex 6PPD from Solutia)  
Zinc oxide: Zinc White No. 3 from Seido Chemical Industry Co., Ltd.

Stearic acid: Beads Stearic Acid from NOF Corporation  
20 Calcium stearate: Calcium stearate from NOF Corporation

**[0034]** [Configuration of Inflation Molding Equipment]

An inflation molding equipment comprising a  $\phi 90$  mm single-screw extruder, a cylindrical die, and an air ring device (manufactured by Macro Engineering & Technology  
25 Inc.) was used to produce a cylindrical film. The cylindrical die had an annular discharge port with an outer diameter of 20.32 cm (8 inches). The annular discharge port of the cylindrical die was directed upward in the vertical direction (that is, the direction  
30 opposite to direction of gravity). A pair of guides and pair of pinch rolls were arranged above the annular discharge port in the vertical direction. A plurality of

- 22 -

guide rolls and a windup roll were arranged so that the cylindrical film extruded from the annular discharge port is folded by the pair of pinch rolls, then the folded cylindrical film is wound up by the windup roll through the plurality of guide rolls. D10S dual lip air ring manufactured by Macro Engineering & Technology Inc. with an inside diameter of 9 inch was used as an air ring device.

[0035] [Molding Conditions]

The cylinder temperature of the extruder was 230°C, and the cylinder temperature of the cylindrical die was 240°C. The molten thermoplastic elastomer composition was shaped at a drawing speed of about 10 m/min so as to give a cylindrical film having a thickness of 0.1 mm and a diameter of 400 mm by a discharge rate of 80 kg/h from the cylindrical die. Cooling of the bubble was carried out by an air-cooling method in which air is blown as a cooling gas to the bubble from the air ring device. The cooling gas flow rate was 1487 m<sup>3</sup>/h, and the temperature of the cooling gas was 5°C.

[0036] [Production of Cylindrical Film]

An inflation molding equipment having the above construction was used to form a thermoplastic elastomer composition into a cylindrical film under the above molding conditions. The bubble stability and dimensional precision when producing a cylindrical film and the shrinkage of the produced film were evaluated by the evaluation criteria described below. The results of evaluation are shown in Table 2 below.

[0037] [Bubble Stability]

At a position 50 cm above the die, a pair of infrared sensors were arranged at opposing positions across the bubble. The distances from the bubble were

- 23 -

measured and the standard deviation  $\sigma$  of the variation thereof was used to evaluate the bubble stability of Examples 1 to 6 and Comparative Examples 1 to 5.

$\sigma$  of 20% or more was rated at "1".

5  $\sigma$  of 10% or more and less than 20% was rated as "2".

$\sigma$  of 5% or more and less than 10% was rated as "3".

$\sigma$  of 1% or more and less than 5% was rated as "4".

$\sigma$  of less than 1% was rated as "5".

[0038] The ratings "3", "4", and "5" are levels where  
10 a member for inner liner could be obtained, but the rating "2" is a level where there was severe wrinkling at the time of winding of the cylindrical film and according the cylindrical film was unsuitable for inner liner applications, or it was difficult to carry out molding  
15 over a long period of time. The rating "1" is a level where the bubble was unstable and accordingly it is difficult to continue molding.

[0039] [Dimensional Precision]

Each of the cylindrical films of Examples 1 to 6 and  
20 Comparative Examples 1 to 5 was cut into five pieces in compliance with the width of the desired tire inner liner immediately after being formed. Each piece was measured for circumferential direction length L1 of the cylindrical film at three locations of both cut ends and  
25 center part. The rating of the dimensional precision was carried out as follows.

The case where there was a cylindrical film having a variation in L1 of three locations of more than 10 mm was rated as "1".

30 The case where there was a cylindrical film having a variation in L1 of three locations of more than 5 mm and 10 mm or less was rated as "2".

- 24 -

The case where there was a cylindrical film having a variation in L1 of three locations of more than 3 mm and 5 mm or less was rated as "3".

5 The case where there was a cylindrical film having a variation in L1 of three locations of more than 2 mm and 3 mm or less was rated as "4".

The case where there was a cylindrical film having a variation in the L1 of three locations of 2 mm or less was rated as "5".

10 [0040] In the case of the ratings "3", "4", and "5", the resulting film can be laminated with the adjacent rubber members to form a tire, but in the case of the rating "3", wrinkles were easily formed in the circumferential direction of the film, and in the case of  
15 the rating "4", wrinkles were occasionally formed. In the case of the rating "5", wrinkles were not formed. In the case of the rating "2", wrinkling was severe and many air pockets were formed between the film and adjacent rubber members, or wrinkles became folded parts, resulting  
20 portions not contacting with the adjacent rubber members, and consequently tire molding is difficult. The rating "1" is the level where almost all wrinkles became folded parts and it is difficult to produce a tire.

[0041] [Shrinkage Factor]

25 Each of the cylindrical films of Examples 1 to 6 and Comparative Examples 1 to 5 was cut to in compliance with the width of the desired tire inner liner immediately after being formed and was measured for circumferential direction length L1. After measurement, the film was hung  
30 in a room and allowed to stand at normal temperature for one week, and then was again measured for circumferential direction length L2 at the same position. The shrinkage factor (%) was determined in accordance with the formula

- 25 -

$100 \times (L1 - L2) / L1$ .

The case where shrinkage factor (%) was 2.0% or more was rated as "1".

5 The case where the shrinkage factor (%) was more than 1.5% and less than 2.0% was rated as "2".

The case where the shrinkage factor (%) was more than 1.0% and less than 1.5% was rated as "3".

The case where the shrinkage factor (%) was more than 0.5% and less than 1.0% was rated as "4".

10 The case where the shrinkage factor (%) was 0.5% or less was rated as "5".

[0042] The higher the shrinkage factor, the lower the dimensional precision. The lower the original dimensional precision and the higher the shrinkage factor, the more  
15 failures of wrinkling or folding of the film when laminating it with the adjacent rubber members.

[0043] Table 2

|             | Rectifying<br>cylinder<br>height H | Bubble<br>stability | Dimensional<br>precision | Shrinkage |
|-------------|------------------------------------|---------------------|--------------------------|-----------|
| Comp. Ex. 1 | 0.3D                               | 1                   | 1                        | 5         |
| Comp. Ex. 2 | 0.4D                               | 2                   | 2                        | 5         |
| Ex. 1       | 0.5D                               | 3                   | 3                        | 5         |
| Ex. 2       | 0.7D                               | 4                   | 4                        | 5         |
| Ex. 3       | 0.8D                               | 5                   | 5                        | 5         |
| Ex. 4       | 1.2D                               | 5                   | 5                        | 5         |
| Ex. 5       | 1.3D                               | 5                   | 4                        | 4         |
| Ex. 6       | 1.5D                               | 5                   | 4                        | 3         |
| Comp. Ex. 3 | 1.6D                               | 5                   | 3                        | 2         |
| Comp. Ex. 4 | 2.0D                               | 5                   | 2                        | 2         |
| Comp. Ex. 5 | 3.0D                               | 5                   | 2                        | 1         |

[0044] Table 2 shows that the inflation molding  
20 equipment of the present invention leads to excellent bubble stability, enables production of a cylindrical film with a high dimensional precision and enables reduction in shrinkage of the resulting cylindrical film.

- 26 -

## Industrial Applicability

[0045] The inflation molding equipment of the present invention can be suitably used for the production of a cylindrical film. Further, the cylindrical film produced using the inflation molding equipment of the present invention can be suitably used as an inner liner of a pneumatic tire for the production of a pneumatic tire.

## Reference Signs List

- [0046] 1. Inflation molding equipment
- 10 Extruder
20. Cylindrical die
21. Annular discharge port
30. Air ring device
33. Lower annular outlet port
- 15 34. Upper annular outlet port
- 34A. Outer circumferential wall
- 40A, 40B. Stabilizing plates
- 50A, 50B. Pinch rolls
60. Windup roll
- 20 D1. Outer diameter of annular discharge port
- D2. Diameter of cylindrical film
- F1. Cylindrical film
- F2. Folded cylindrical film

- 27 -

CLAIMS

Claim 1. Inflation molding equipment comprising:  
5 an extruder for melt extruding a stock material,  
a single-layer or multi-layer cylindrical die  
attached to a discharge port of the extruder, and  
an air ring device for cooling a cylindrical film  
extruded from the cylindrical die, wherein  
10 the cylindrical die has an annular discharge port  
having an outer diameter of D1,  
the air ring device is attached on the cylindrical  
die, and  
the air ring device has at least one cooling gas  
15 outlet port, the cooling gas outlet port having a cooling  
gas channel defined by an outer circumferential wall  
having a height of H and an inner circumferential wall  
having a height lower than the outer circumferential  
wall, and  
20 D1 and H satisfy a relationship of the following  
formula (1):

$$0.5 \times D1 \leq H \leq 1.5 \times D1 \quad (1).$$

Claim 2. The inflation molding equipment according  
to claim 1, wherein said stock material is a  
25 thermoplastic elastomer composition having an island-in-  
sea structure comprised of a continuous phase comprising  
a thermoplastic resin and a disperse phase comprising an  
elastomer.

Claim 3. The inflation molding equipment according  
30 to claim 1, wherein D1 and H satisfy a relationship of  
the following formula (2):

$$0.8 \times D1 \leq H \leq 1.2 \times D1 \quad (2)$$

Claim 4. The inflation molding equipment according

- 28 -

to any one of claims 1 to 3 wherein a ratio of an effective length L of a screw to a screw diameter D of the extruder,  $L/D$ , is 32 or more.

5           Claim 5. The inflation molding equipment according to any one of claims 1 to 4, wherein the air ring device does not have a rectifying cylinder spaced apart from the outer circumferential wall of the cooling gas outlet port at the outer circumferential side of the outer circumferential wall.

10           Claim 6. A process for producing a cylindrical film, comprising:

          providing the inflation molding equipment according to any one of claims 1 to 5;

          melt extruding a molten thermoplastic elastomer  
15       composition from the extruder to the cylindrical die, the molten thermoplastic elastomer composition comprising a thermoplastic resin selected from the group consisting of a polyamide-based resin, a polyvinyl-based resin, a polyester-based resin, and a blend of two or more of  
20       these resins as a continuous phase, and a modified elastomer as a disperse phase; and

          extruding the molten thermoplastic elastomer composition as a cylindrical film from the cylindrical die.

25           Claim 7. A process for producing a pneumatic tire comprising:

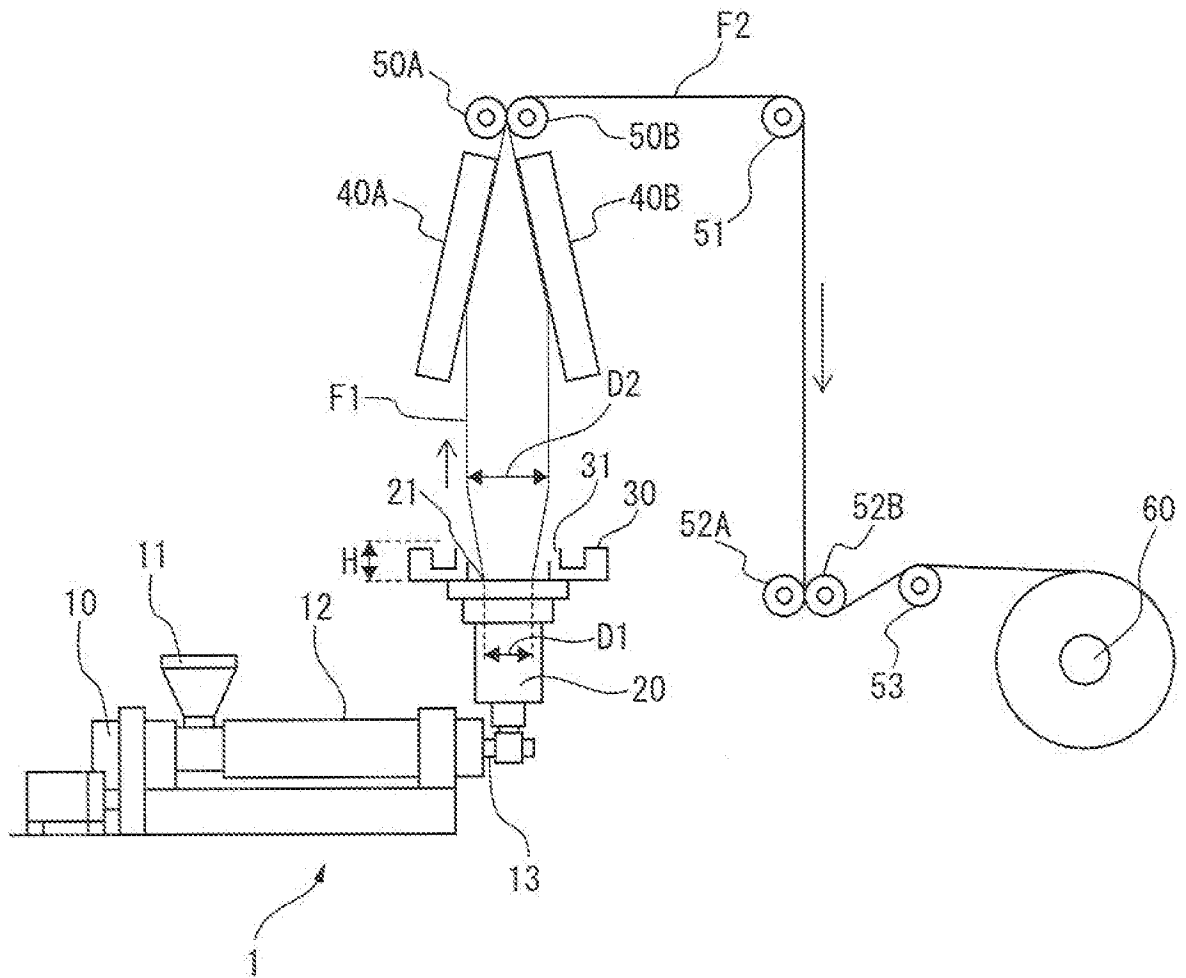
          producing a cylindrical film by the process according to claim 6;

          cutting the resulting film in accordance with a size  
30       of the pneumatic tire to form an inner liner;

          forming a green tire comprising the inner liner; and  
          vulcanizing the green tire to form a pneumatic tire.

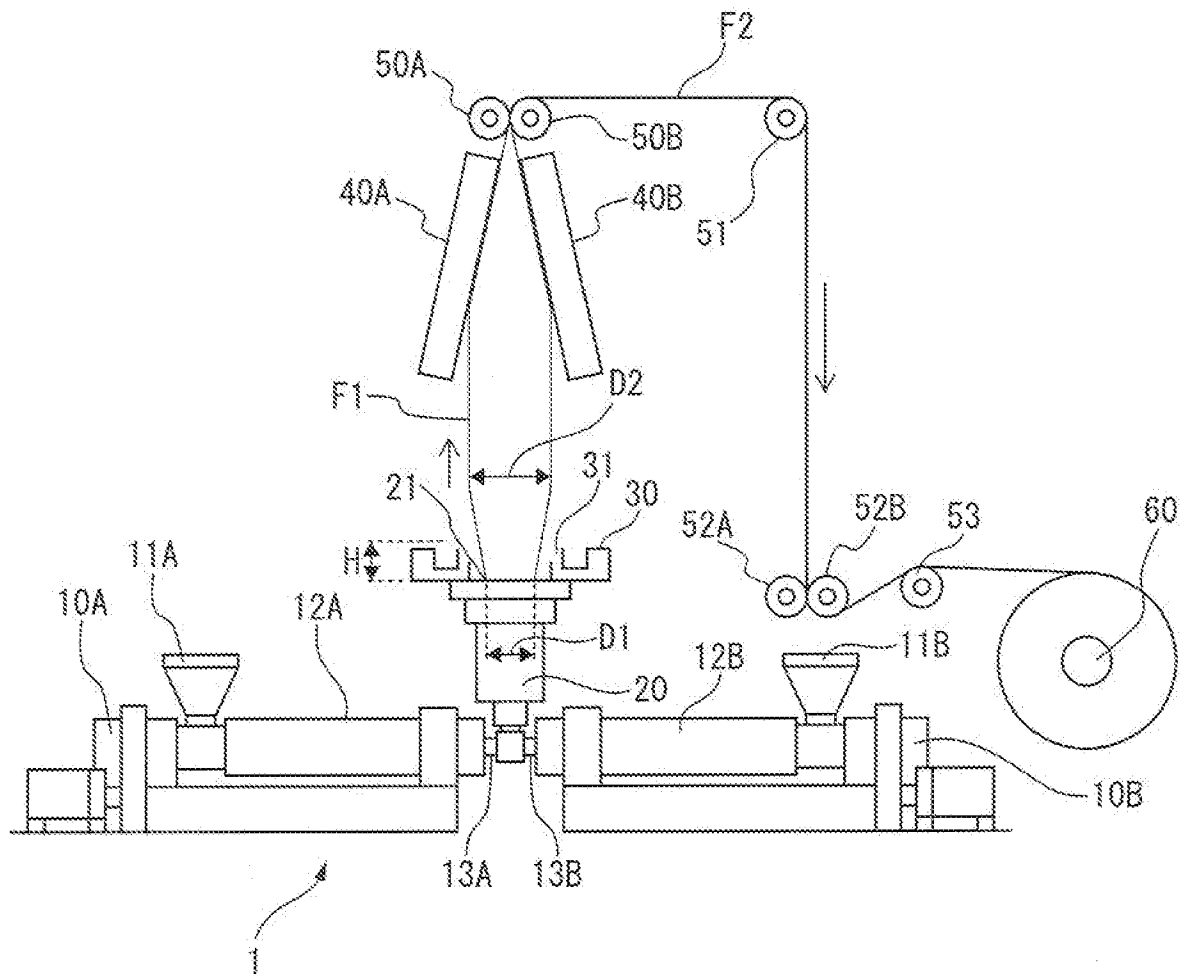
1/3

FIG. 1



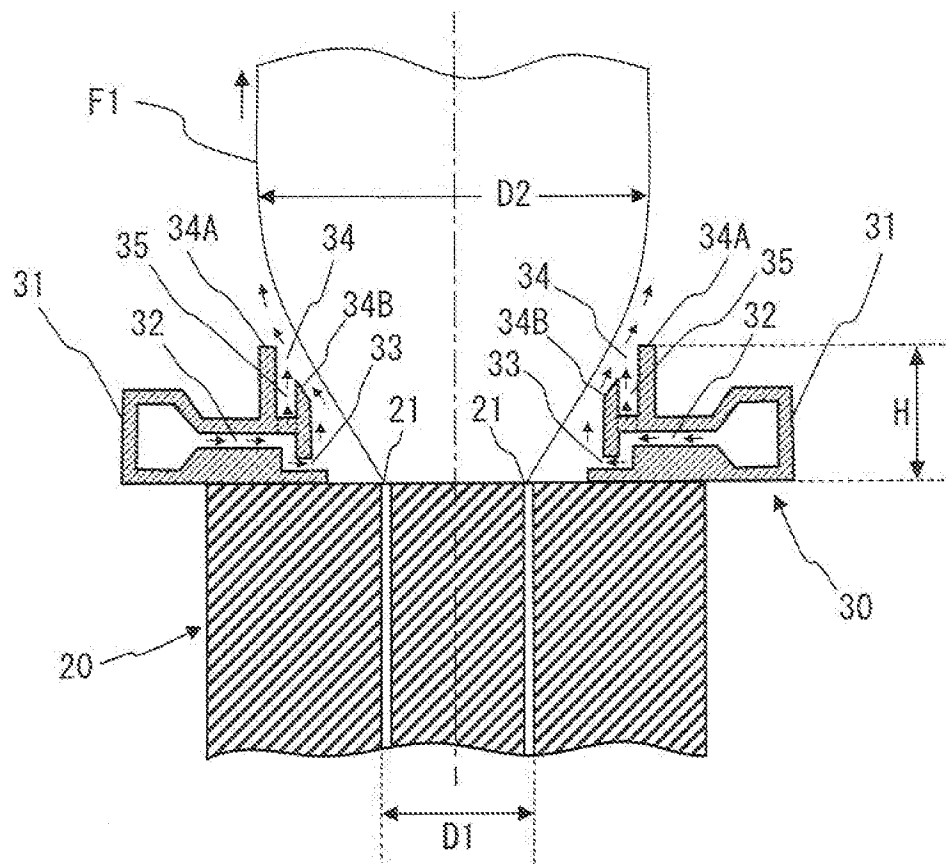
2/3

FIG. 2



3/3

FIG. 3



## INTERNATIONAL SEARCH REPORT

International application No.

PCT/US16/17288

## A. CLASSIFICATION OF SUBJECT MATTER

IPC(8) - B29C 55/28, 49/04 (2016.01)

CPC - B29C 55/28, 49/04

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC(8): B29C 55/28, 49/04, 49/28, 49/64 (2016.01)

CPC: B29C 55/28, 49/04, 49/28, 49/64, 49/6427

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

PatSeer (US, EP, WO, JP, DE, GB, CN, FR, KR, ES, AU, IN, CA, INPADOC Data); EBSCO; IEEE.com; Google Scholar; Google Patent; film, thermoplastic, elastomer, inflation, blown, bubble, extrusion, extrude, die, diameter, cooling, air ring, channel, height, length, H/D, L/D

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

| Category* | Citation of document, with indication, where appropriate, of the relevant passages                                                                                                    | Relevant to claim No. |
|-----------|---------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|-----------------------|
| Y         | US 5,468,444 A (YAZAKI, T et al) 21 November 1995; figures 3-4, 6-7; column 1, lines 27-32; column 2, lines 45-59; column 7, lines 34-67; column 8, lines 1-20; column 10, lines 1-30 | 1-3, 4/1-3            |
| Y         | WO 2015/183577 A1 (EXXONMOBIL CHEMICAL PATENTS INC) 03 December 2015; figure 1B; paragraphs [0021], [0026]                                                                            | 1-3, 4/1-3            |
| Y         | US 2014/0150947 A1 (SHANNON, PC et al) 05 June 2014; claims 1-2                                                                                                                       | 2, 4/2                |
| Y         | US 2013/0295364 A1 (SAAVEDRA, JV et al) 07 November 2013; paragraph [0047]                                                                                                            | 4/1-3                 |
| A         | US 2004/0113331 A1 (MIYATA, T et al) 17 June 2004; entire document                                                                                                                    | 1-3, 4/1-3            |
| A         | US 5,795,533 A (MEHNERT, G) 18 August 1998; entire document                                                                                                                           | 1-3, 4/1-3            |
| A         | US 4,643,657 A (ACHELPOHL, F et al) 17 February 1987; entire document                                                                                                                 | 1-3, 4/1-3            |



Further documents are listed in the continuation of Box C.



See patent family annex.

\* Special categories of cited documents:

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier application or patent but published on or after the international filing date

"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art

"&amp;" document member of the same patent family

Date of the actual completion of the international search

22 March 2016 (22.03.2016)

Date of mailing of the international search report

22 APR 2016

Name and mailing address of the ISA/

Mail Stop PCT, Attn: ISA/US, Commissioner for Patents

P.O. Box 1450, Alexandria, Virginia 22313-1450

Facsimile No. 571-273-8300

Authorized officer

Shane Thomas

PCT Helpdesk: 571-272-4300

PCT OSP: 571-272-7774

# INTERNATIONAL SEARCH REPORT

International application No.

PCT/US16/17288

## Box No. II Observations where certain claims were found unsearchable (Continuation of item 2 of first sheet)

This international search report has not been established in respect of certain claims under Article 17(2)(a) for the following reasons:

1. ☐ Claims Nos.:  
because they relate to subject matter not required to be searched by this Authority, namely:
  
2. ☐ Claims Nos.:  
because they relate to parts of the international application that do not comply with the prescribed requirements to such an extent that no meaningful international search can be carried out, specifically:
  
3. ☒ Claims Nos.: 5-7  
because they are dependent claims and are not drafted in accordance with the second and third sentences of Rule 6.4(a).

## Box No. III Observations where unity of invention is lacking (Continuation of item 3 of first sheet)

This International Searching Authority found multiple inventions in this international application, as follows:

1. ☐ As all required additional search fees were timely paid by the applicant, this international search report covers all searchable claims.
2. ☐ As all searchable claims could be searched without effort justifying additional fees, this Authority did not invite payment of additional fees.
3. ☐ As only some of the required additional search fees were timely paid by the applicant, this international search report covers only those claims for which fees were paid, specifically claims Nos.:
  
4. ☐ No required additional search fees were timely paid by the applicant. Consequently, this international search report is restricted to the invention first mentioned in the claims; it is covered by claims Nos.:

### Remark on Protest

- ☐ The additional search fees were accompanied by the applicant's protest and, where applicable, the payment of a protest fee.
- ☐ The additional search fees were accompanied by the applicant's protest but the applicable protest fee was not paid within the time limit specified in the invitation.
- ☐ No protest accompanied the payment of additional search fees.