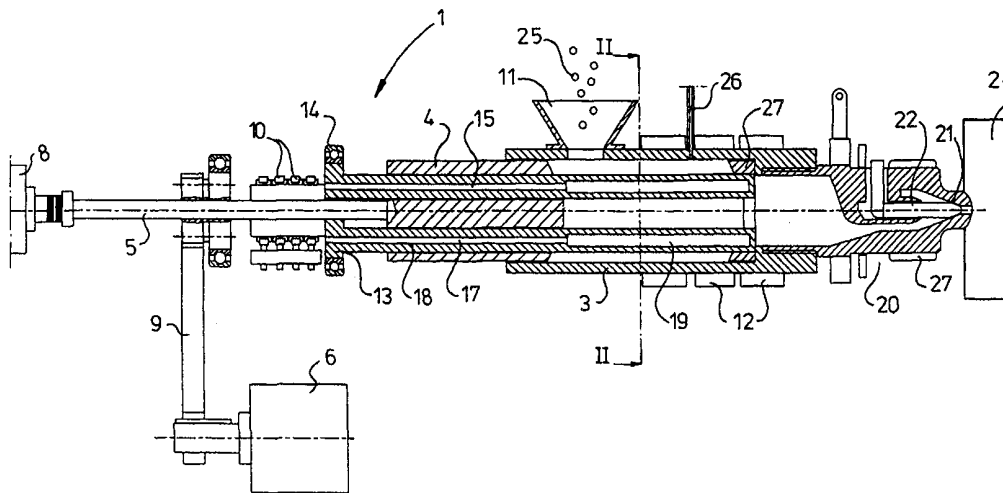




INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

<p>(51) International Patent Classification <sup>7</sup> : <b>B29C 45/53, 45/58, B01F 15/02</b></p>	<p><b>A1</b></p>	<p>(11) International Publication Number: <b>WO 00/02718</b> (43) International Publication Date: 20 January 2000 (20.01.00)</p>
<p>(21) International Application Number: PCT/NL99/00447 (22) International Filing Date: 13 July 1999 (13.07.99) (30) Priority Data: 1009627 13 July 1998 (13.07.98) NL (71) Applicant (for all designated States except US): TECHNISCHE UNIVERSITEIT DELFT [NL/NL]; P.O. Box 5058, NL-2600 GB Delft (NL). (72) Inventors; and (75) Inventors/Applicants (for US only): BEUKERS, Adriaan [NL/NL]; A. Thijm laan 17, NL-2106 EG Heemstede (NL). WILTINK, Frederik, Johan [NL/NL]; Lambrecht van Dalelaan 86, NL-2015 EV Haarlem (NL). VAN BREUGEL, Josephus, Hendrikus [NL/NL]; Berliozstraat 17, NL-5283 LA Boxtel (NL). (74) Agent: DE BRUIJN, Leendert, C.; Nederlandsch Octrooibureau, Scheveningseweg 82, P.O. Box 29720, NL-2502 LS The Hague (NL).</p>		<p>(81) Designated States: AE, AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, CA, CH, CN, CU, CZ, DE, DK, EE, ES, FI, GB, GD, GE, GH, GM, HR, HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, UA, UG, US, UZ, VN, YU, ZA, ZW, ARIPO patent (GH, GM, KE, LS, MW, SD, SL, SZ, UG, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, GW, ML, MR, NE, SN, TD, TG).</p> <p><b>Published</b> With international search report. In English translation (filed in Dutch).</p>

(54) Title: DEVICE AND METHOD FOR THE PREPARATION OF A MIXTURE COMPRISING FIBRE-REINFORCED THERMOPLASTIC PELLETS



(57) Abstract

This material must be plasticised and in order to be able to process such a mixture by injection moulding or flow moulding it is necessary for, on the one hand, a rise in pressure and, on the other hand, mixing of the material to take place by means of the device. In order as far as possible to prevent damage to the fibres during a mixing, it is proposed to effect mixing using a piston/cylinder device (3, 4) in which a stirrer construction is fitted, consisting of a number of mixing rods (15, 16, 17) which rotate together with the piston (4). With this arrangement the mixing rods (15, 16, 17) essentially cover the entire surface of the volume to be mixed.

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Device and method for the preparation of a mixture comprising fibre-reinforced thermoplastic pellets

The present invention relates to a device for the preparation of a mixture according to the preamble of Claim 1. A device of this type is disclosed in US-A 2 990 978. This publication describes a mixing device for foodstuffs with a paste consistency. The latter have relatively low viscosity.

It is known to process thermoplastic and thermosetting materials by, for example, injection moulding or flow moulding. These techniques are reasonably developed.

However, in order to increase the strength of the articles made of plastic material which are produced in this way, it has been proposed to use thermoplastic fibre-reinforced plastics. The use of a thermoplastic material compared with a thermosetting material has the advantage that the cycle times of injection moulding can be shortened, the impact strength increases, a good mix of rigidity and strength is obtained, more particularly an adequate elongation at break, and there is a greater freedom of choice of material, whilst, finally, recycling is possible.

However, it has been found that wetting of the fibre in the thermoplastic matrix constitutes a problem. Solutions to this have been proposed in the prior art, for example by producing fibre-reinforced thermoplastics with the aid of powder impregnation. These thermoplastics are supplied in pellet form. The pellets obtained are cut or chopped from a strip. Such pellets are fed in the conventional manner to the feed device for a processing device, such as an injection moulding machine. Such a feed device consists of a screw extruder. In the latter, on the one hand, the pressure is raised and, on the other hand, optimum mixing of the material is obtained. The disadvantage is, however, that the screw of the screw extruder damages the fibres or gives rise to fibre degradation. Moreover, such extruders are very expensive. Because fibre degradation takes place, the mechanical properties of the product obtained by, for example, injection moulding are impaired and because of the high costs economic production of such products is not possible. In practice it is not possible to process more than 30 % by volume fibre with such expensive machines. On the other hand, there is certainly a need for such products containing fibres and thus having higher strength, more particularly to replace a wide variety of components made of aluminium and the like. Use of plastic has the advantage that, in particular, the weight of the construction concerned can be reduced, which, for example, in the case of vehicles has the result that the fuel

consumption thereof can fall.

US-A 3 844 699 describes a press device for simultaneous mixing and injection moulding of polymer materials. In the piston/cylinder assembly used in this device there is a stirrer rod arranged in the middle which extends only over a limited portion of the surface to  
5 be stirred.

The aim of the present invention is to provide a device for the preparation of a mixture comprising fibre-reinforced thermoplastic or thermosetting pellets or reinforced rubber particles. This fibre material can comprise any material known in the prior art, such as carbon, aramide and glass. The same applies in respect of the thermoplastic, thermosetting or  
10 rubber materials.

A device of this type must, on the one hand, be capable of adequately plasticising and pressurising such a material and, on the other hand, have a relatively low cost price, whilst at most only slight fibre degradation may take place during plasticising.

This aim is realised with a device as described above having the characterising features  
15 of Claim 1.

Because the mixing rods are located different distances away from the axis of rotation of the piston, intensive mixing can be guaranteed. Preferably, the number of rods is so chosen and the rods are each so mounted that the entire mixing region is covered by the rods. It has been found that, where thermoplastic materials into which large quantities of fibres have to be  
20 mixed are used, the mixture is not mixed sufficiently thoroughly to become homogeneous if a mixing device according to US-A 2 990 978 is used. The device according to US-A 2 990 978 is not intended for mixing fibre-containing materials. It must be understood that the percentage of fibre in such materials can be particularly high. A value of 50 % by volume fibre may be mentioned by way of example, it being possible for the fibres to be of  
25 appreciable length, for example longer than 1 cm. It must be understood that the differing distances from the stirrer rods to the axis of rotation can also be obtained by making the rods of different diameter. The techniques known from the prior art for covering the entire surface to be mixed with the aid of the mixing device also fall under the present invention.

As a result of the use of a piston/cylinder assembly, damage to the fibres is largely  
30 prevented, that is to say no or hardly any fibre degradation takes place. However, because a piston/cylinder assembly does not ensure adequate mixing, it is necessary to use a separate mixing device which according to the invention consists of mixing means that can be rotated with the piston. Such mixing means can comprise any means known in the prior art.

According to the invention, the mixing means consist of one or more mixing rods which extend through the piston in the path of movement of the piston. That is to say the mixing rod or rods execute a, for example rotary, movement together with the piston. The piston consequently makes a reciprocal movement and a rotary movement. Such a rotary movement on the one hand provides for good mixing, so that a uniform distribution of heat over the surface of the cylinder is obtained, and, on the other hand, imposes no stress on the fibres, so that fibre degradation is negligible.

In order to obtain a homogeneous temperature over the cross-section of the cylinder it is possible, as a first step, to fit a number of heating elements on the outer circumference of the cylinder. The temperature of the thermoplastic material provided with fibre material is increased by this means. A further homogenisation of temperature can be obtained if the stirrer rods are provided with heating elements. Such heating elements can comprise heating elements operating with fluid and can also be electrical heating elements. It will be understood that suitable adjustment and control means must be fitted in order to control the temperature of the various heating elements.

Mixing with the aid of the mixing rods described above is promoted by fitting different mixing rods. If the mixing rods execute a rotary movement together with the piston about the axis of the piston, optimum homogeneous mixing can be obtained if the distance of the various mixing rods from the axis of rotation of the piston is different for each rod. That is to say as wide as possible an area of the surface of the cylinder is covered by the mixing rods.

Further optimisation of the mixing is obtained if one or more of the mixing rods fitted is provided with a surface relief. Said relief can be ribs, grooves and the like.

In order to optimise the conditions during mixing and plasticising and in particular to prevent oxidation of either the plastic or the fibres at the high temperature at which said plasticising takes place, according to the invention a gas feed is provided in which an active, inert or oxidation-preventing gas can be introduced. An example is ozone or nitrogen.

The invention also relates to an injection moulding assembly or other processing assembly, comprising an injection moulding device or other processing device having at least two mould halves, which delimit a mould cavity and are movable with respect to one another, and a feed for plastic, as well as at least one device for preparing a mixture of fibre-reinforced plastics in accordance with what has been described above, connected to said feed for plastic.

If the pressure generated with the device described above for the preparation of a mixture comprising fibre-reinforced thermoplastic pellets, and more particularly by the

piston/cylinder assembly, is inadequate, it is possible, according to the invention, to provide for a further pressure-raising device to be installed between the device and the processing device. This further device can be a further piston/cylinder device.

5 The aim must, of course, be to match the capacity of the device for mixing or plasticising the fibre-reinforced thermoplastic plastic material as far as possible to the processing device. However, it is possible that the capacity of an device for the preparation of such a plastic mixture is inadequate. In such a case several such devices can be connected to a processing device and successively dispense a quantity of material.

10 It is possible for the piston of the piston/cylinder assembly to execute a complete stroke for each shot of the processing device. It is also possible that the volume of the piston/cylinder assembly is greater than the volume required to fill the injection moulding machine once.

The invention also relates to a method for the preparation of a mixture consisting of plastic and fibre or other filler material, wherein the plastic is introduced with the fibres or other filler material into a mixing chamber and is placed under pressure while mixing by stirring. With this method stirring is preferably carried out using mixing rods which essentially cover the entire volume to be stirred. As indicated above, the pressure in the mixture is preferably generated with the aid of a piston/cylinder assembly. The feed of the plastic containing fibres or other filler material is preferably achieved by pellets in which the fibres or filler material concerned are present. If fibres are used, these can be of an appreciable length. Preferably, said fibres are at least 1 cm long and more particularly the length thereof is between 10 and 100 mm. According to the invention, preferably 40 % by volume fibre is used. A mixture of this type containing such a high proportion by volume of fibre is particularly difficult to mix, but with the measures described above it has been found, surprisingly, that the various aspects can be achieved. Very strong plastics can be obtained in this way. Compared with a conventional steel construction, it is possible to produce a more rigid and lightweight construction. Furthermore, the invention makes it possible to produce products relatively inexpensively.

30 The invention will be explained in more detail below with reference to an illustrative embodiment shown in the drawing. In the drawing:

Fig. 1 shows, diagrammatically and partially in longitudinal section, the plasticising device according to the invention; and

Fig. 2 shows a section along the line II-II in Fig. 1.

In Fig. 1 the device according to the invention for mixing and plasticising fibre-reinforced thermoplastic plastic pellets is indicated in its entirety by 1. This device is coupled to an injection moulding device, which is indicated highly diagrammatically by 2. Details of the injection moulding device are not given because the device can be any injection moulding  
5 device having one or more movable mould halves known from the prior art. It must be understood that this can be combined with any other processing device known from the prior art.

The device for plasticising thermoplastic plastic material that is fibre-reinforced comprises a cylinder 3 in which a piston 4 is reciprocatingly mounted. A piston rod 5 is  
10 connected to the piston 4, which piston rod 5 engages via a coupling 7 on a hydraulic cylinder. Said coupling is a coupling that absorbs pressure and permits rotation. That is to say, piston rod 5 is able to rotate relative to the cylinder of the hydraulic cylinder 8.

A motor which is able to bring piston rod 5 into rotation via a belt 9 is indicated by 6. Piston 4 is rotated by this means. A carrier plate 13, on which a number of, in this case three,  
15 stirrer rods 15-17 are mounted, is also brought into rotation by this means. These stirrer rods can be provided with a surface relief. Carrier plate 13 is supported on a housing, which is not shown in more detail, via a bearing 14. Sliding contacts 10 are present, which provide electrical contact between the surroundings and each of the stirrer rods 15-17 and more particularly heating elements 19 mounted therein. A carrier plate mounted in a sliding bearing  
20 is indicated by 27, the other ends of the rods 15-17 bearing in said carrier plate.

In the embodiment shown, piston 4 is provided with three openings for accommodating the stirrer rods 15-17. Piston 4 is able to move to and fro along each of said stirrer rods 15-17. The stirrer rods are not arranged around a circle concentrically with the piston. As can be seen  
25 from Fig. 2, the distances a, b and c between the stirrer rods 15-17 and the axis of rotation of the piston are different. As a result the major proportion of the cross-sectional surface of the cylinder is covered on rotation.

The head of the device according the invention is indicated by 20 in Fig 1. This head is provided with a conventional valve seat 21 in which a displaceable needle 22 is fitted. Some form of valve in the device according to the invention is important to be able to build up  
30 pressure. The cylinder 3 is provided with various heating elements 12, whilst the head is provided with a heating element 27. The feed of plastic material takes place via the fill opening or funnel 11 and the pellets to be introduced therein are indicated by 25. A gas feed 26 is also fitted for feeding in an inert or active gas.

The device described above functions as follows. The starting position is that in which the piston 4 is in the position in which it has been brought to the left, in which position pellets 25, such as thermoplastic pellets reinforced with carbon fibres, are introduced into the cylinder chamber at a relatively low temperature. Piston 4 is then moved to the right while  
5 supplying an inert or active gas through gas feed 26. During this movement the piston, and thus the stirrer rods 15, rotate. A speed of revolution of 60 revolutions per minute may be mentioned by way of example. It will be understood that the invention is, however, not restricted to this. As a result of the presence of the heating elements and as a result of the compression of the plastic material, a rise in temperature to, for example, 400°C takes place.  
10 If the pressure and temperature have reached the desired value, needle 21 is moved to the left and the plastic material is transported to the injection moulding device, where further processing takes place.

It has been found that with the device described above no or hardly any damage occurs to the fibres which are present in the pellets and that material is obtained which has  
15 characteristics which are essentially the same in all directions. Using this device it is possible to bring materials having a high fibre content, for example 50-60 % by wt fibre, into an injection moulding device at sufficiently high temperature and without substantial damage to the fibres. Using the device described above it is possible to achieve a pressure of approximately 50 atmospheres in the plastic without any special measures. If such a pressure  
20 is inadequate, special measures then have to be taken, such as connecting a pressure-raising device between device 1 and injection moulding device 2.

The materials obtained using the device and the method according to the invention are compared with known materials below. In the following table sheet material from Azdel (so-called GMT sheet) is compared with two materials according to the invention: PA6-LCF and  
25 PA12-LCF. The first material mentioned is nylon 6 reinforced with 12.5 mm long carbon fibres. The second material is nylon 12 reinforced with 12.5 mm long carbon fibres. Finally, these materials are compared with the conventional construction materials aluminium and steel.



		Azdel PM1040D	PA6- LCF	PA12- LCF	Aluminium	Steel
Fibre load	[% by wt]	40	28	30	-	-
Fibre volume	[% by volume]	20	20	20	-	-
Density	[g/cm <sup>3</sup> ]	1.21	1.26	1.15	2.8	7.8
Tensile strength	[Mpa]	100	150	200	160	360-540
Young's modulus	[Gpa]	6	15-20	20-25	72	210
Specific modulus e/p	[MNm/g]	5	14	20	25.7	26.9
Material costs	[US\$/kg]	2.75	15	20	1.96	0.74
Stiffness-specific costs	[US\$/GNm]	0.55	1.07	1	0.08	0.03

It can be seen from this comparison that the materials processed using the method and device according to the invention have particularly good properties, in which context it is pointed out that, in principle, any shape can be produced by this means. This is in contrast to the sheet material that is described in the left-hand column of the table. With a sheet material  
5 of this type, plastic articles are currently made by pressing, thickening being achieved by placing various parts of sheet material on top of one another. This method is laborious and incurs loss, whilst, moreover, finishing of the products is necessary. These problems do not exist with the proposal according to the invention.

Although the invention has been described above with reference to the use of ready-to-  
10 use pellets, it is possible for users to prepare specific mixtures themselves by feeding the various components or unfilled plastic granules or thermopowder individually or simultaneously.

It will also be understood that the mixing rods do not necessarily have to be constructed absolutely parallel to the axis of movement of the piston. The various rods can be constructed  
15 in spiral form or in accordance with another curve. The only essential aspect for the invention is that a separate stirrer is present inside the cylinder and that, in order to prevent damage to the fibres, the combination that is implemented with screw extruders is not used.

A number of modifications to various components of the device described above are immediately obvious to those skilled in the art. Such modifications are considered to fall  
20 within the scope of the appended claims.

Claims

1. Device (1) for the preparation of a mixture, comprising fibre-reinforced pellets (25), having a piston (4)/cylinder (3) assembly, comprising a mixing device, mixing means  
5 (15, 16, 17) which can be rotated with said piston, said mixing means comprise mixing rods (15, 16) extending along the movement path of said piston, characterised in that at least two mixing rods are fitted and the distance (a, b, c) of each of said rods from the axis of rotation of said piston is different for each rod.
2. Device according to Claim 1, wherein said mixing means are provided with  
10 heating means (19).
3. Device according to one of the preceding claims, wherein said rods are provided with a surface relief.
4. Device according to one of the preceding claims, comprising a gas feed (26).
5. Processing assembly, comprising a processing device (2) having at least two  
15 mould halves, which delimit a mould cavity and are movable with respect to one another, and a feed for plastic, as well as at least one device for preparing a mixture of fibre-reinforced plastics according to one of the preceding claims connected to said feed for plastic.
6. Assembly according to Claim 5, wherein a device which raises the pressure of the  
20 plastic to be injection moulded is arranged between said device for the preparation of a mixture of fibre-reinforced thermoplastic material and said feed for plastic.
7. Assembly according to Claim 5 or 6, comprising a number of devices (1) according to one of Claims 1 - 4, connected to a common feed for plastic for said injection moulding device (2).
8. Method for the preparation of a mixture consisting of plastic and fibres or other  
25 filler material, wherein the plastic is introduced with the fibres or other filler material into a mixing chamber and is brought under pressure while mixing by stirring.
9. Method according to Claim 8, wherein stirring is carried out by means of mixing rods which essentially cover the entire volume to be stirred.
10. Method according to Claim 8 or 9, wherein bringing under pressure comprises  
30 pressing using a piston/cylinder assembly.
11. Method according to one of Claims 8 - 10, wherein the feed of plastic and fibres comprises the feed of plastic pellets containing fibres.
12. Method according to one of Claims 8 - 11, wherein the mixture contains at least

40 % by volume fibre.

13. Method according to one of Claims 8 - 12, wherein at least some of the fibres are at least 1 cm long.

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fig-1

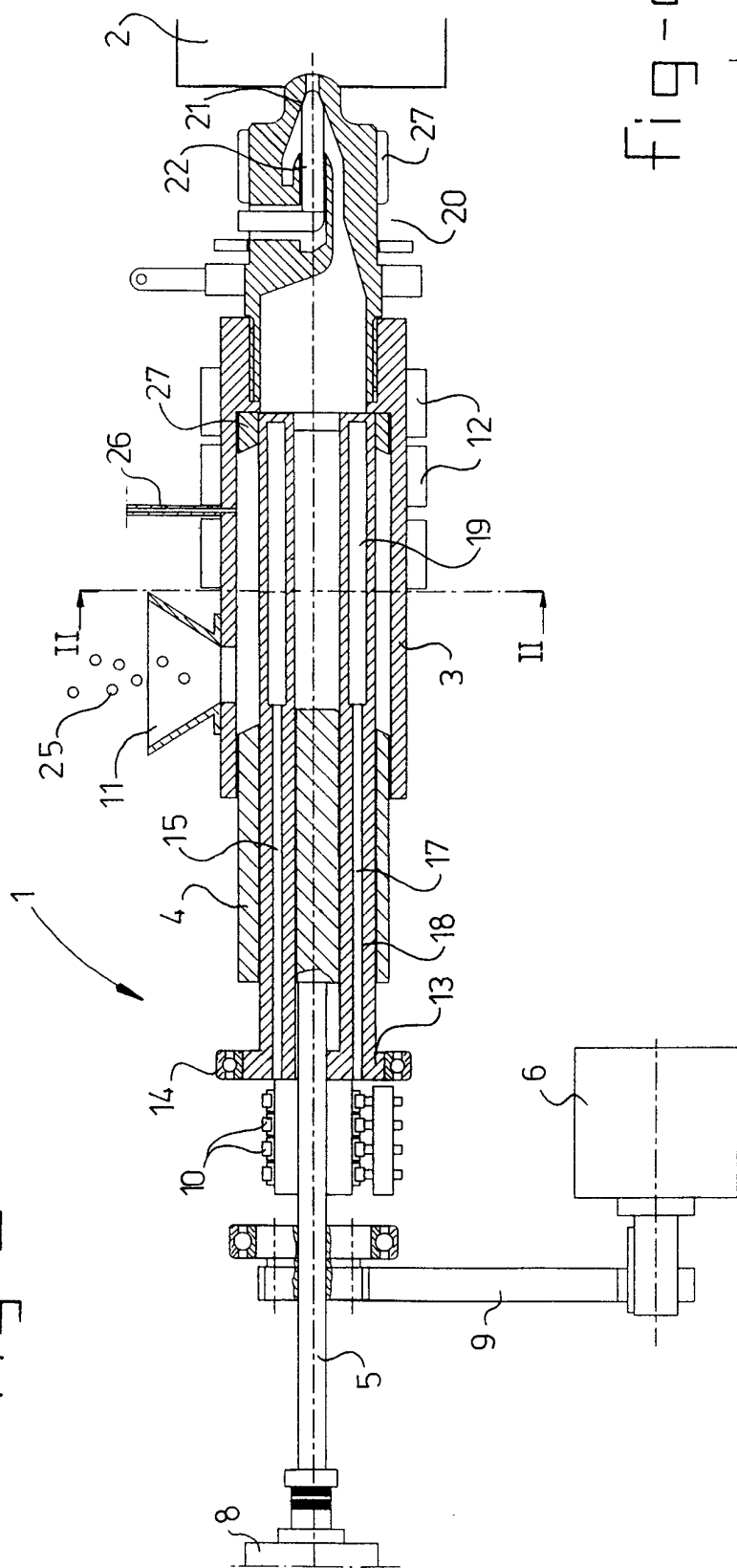
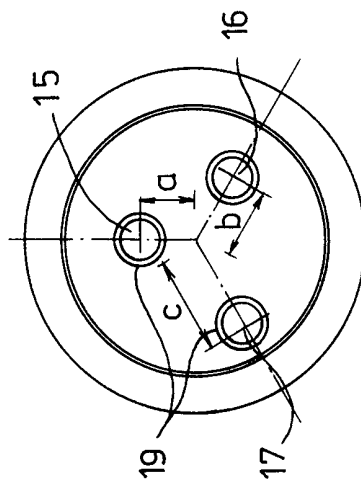


fig-2



# INTERNATIONAL SEARCH REPORT

International Application No  
PCT/NL 99/00447

<b>A. CLASSIFICATION OF SUBJECT MATTER</b> IPC 7 B29C45/53 B29C45/58 B01F15/02				
According to International Patent Classification (IPC) or to both national classification and IPC				
<b>B. FIELDS SEARCHED</b>				
Minimum documentation searched (classification system followed by classification symbols) IPC 7 B29C B01F				
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 practical, search terms used)				
<b>C. DOCUMENTS CONSIDERED TO BE RELEVANT</b>				
Category °	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.		
A	US 2 990 978 A (P. CHAROS) 4 July 1961 (1961-07-04) cited in the application the whole document ---	1,3		
A	US 3 844 699 A (MAXWELL B) 29 October 1974 (1974-10-29) cited in the application column 4, line 56 - line 67; figures 2-7 column 7, paragraph 3 ---	1,5		
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<input checked="" type="checkbox"/> Further documents are listed in the continuation of box C.				
<input checked="" type="checkbox"/> Patent family members are listed in annex.				
° Special categories of cited documents :				
<table style="width: 100%; border: none;"> <tr> <td style="width: 50%; border: none; vertical-align: top;">                     "A" document defining the general state of the art which is not considered to be of particular relevance                      "E" earlier document 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                 </td> <td style="width: 50%; border: none; vertical-align: top;">                     "I" 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                 </td> </tr> </table>			"A" document defining the general state of the art which is not considered to be of particular relevance "E" earlier document 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	"I" 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. "&" document member of the same patent family
"A" document defining the general state of the art which is not considered to be of particular relevance "E" earlier document 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	"I" 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. "&" document member of the same patent family			
Date of the actual completion of the international search	Date of mailing of the international search report			
20 October 1999	29/10/1999			
Name and mailing address of the ISA European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Tx. 31 651 epo nl, Fax: (+31-70) 340-3016	Authorized officer  Bollen, J			

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International Application No  
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C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT		
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Information on patent family members

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			SE 8604237	07-04-1988
			US 4909724	20-03-1990
			US 4842507	27-06-1989
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