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(54) Title: PREPOLYMER, POLYOL COMPOSITION AND PROCESS FOR MAKING A FLEXIBLE FOAM

(57) Abstract: Isocyanate-terminated prepolymer having an NCO-value of 5-30% by weight and being the reaction product of an excessive amount of diphenylmethane diisocyanate comprising at least 80% by weight of 4,4'-diphenylmethane diisocyanate and a polyoxyethylene-polyoxypropylene polyol having an average molecular weight of 2000-10000, an average nominal hydroxy functionality of 2-6, an oxyethylene content of 21-45% by weight and a structure of the type -PO-PO/EO-EO wherein the PO block comprises 60-90% of the PO and the ratio of tipped EO : random is 3:1 to 1:3, and a process for preparing a flexible foam by reacting in a mould and at an index of 70-120, the above prepolymer or a composition comprising this prepolymer and b 1) a polyoxyethylene-polyoxypropylene polyol, having an average nominal hydroxy functionality of 2-6, and a oxyethylene (EO) content of more than 50% by weight; and optionally 2) a polyoxyethylene-polyoxypropylene polyol, having an average nominal hydroxy functionality of 2-6, an EO content of between 20-50% by weight and a primary hydroxy content of at least 50%, calculated on the number of primary and secondary hydroxyl groups; and c) water; and optionally d) additives and auxiliaries known per se.

PREPOLYMER, POLYOL COMPOSITION AND PROCESS FOR MAKING A FLEXIBLE FOAM

The present invention is concerned with a prepolymer, a polyol composition and a process for preparing a flexible polyurethane foam.

In WO 01/32735 a process has been described for such a foam using a prepolymer having an NCO value below 20% by weight. Such a process has as disadvantages that an extra production step is needed to make the prepolymer and that such prepolymers have often a higher viscosity which may hamper the easiness to make the foams. Still further the hardness of the foams is not high enough for foams having a relatively low hard-block content and a relatively high density.

In WO 01/60884 a process for preparing a flexible polyurethane foam is disclosed using a high amount of a polyol having a high oxyethylene content and a high primary hydroxyl content together with a polyol with a medium oxyethylene content; see e.g. example 8. When the experiment was repeated in a closed mould no satisfactory foam was obtained.

WO 01/32736 discloses a process for preparing foams using similar polyols together with a polyol having a low oxyethylene content. The process is a one shot process or a prepolymer process wherein the prepolymer has a high NCO value. The foams obtained have visco-elastic properties.

US 5594097 and US 4559366 disclose the use of polyols having an intermediate amount of oxyethylene groups, like from 20-50% by weight, in preparing flexible foams. US5459170 discloses prepolymers from such polyols.

Surprisingly we now have found that flexible polyurethane foams with a better hardness / density / hard-block-content relation are obtainable with good further properties, like strength, compression set, creep properties, resilience, vibration transmissibility, elongation and a nice feel, by the following easily conductable process by using a special prepolymer and a polyoxyethylene polyoxypropylene polyol with a certain high oxyethylene content and optionally a polyol with a certain medium oxyethylene content and optionally a specific polyoxyethylene polyol.

Therefore the present invention is concerned with a novel prepolymer, a composition of other polyisocyanates and this novel prepolymer and a process for making flexible polyurethane foams by reacting this novel prepolymer or composition with a polyol and water.

The prepolymer according to the present invention is an isocyanate-terminated prepolymer having an NCO-value of 5-30% by weight and preferably 5-19% by weight and being the reaction product of an excessive amount of diphenylmethane diisocyanate (MDI) comprising at least 80 and preferably at least 90% by weight of 4,4'-diphenylmethane diisocyanate and a polyoxyethylene-polyoxypropylene polyol having an average molecular weight of 2000-10000, an average nominal hydroxy functionality of 2-6, an oxyethylene content of 21-45% by weight and a structure of the type -PO-PO/EO-EO wherein the (first) PO block comprises 60-90% of the PO and the ratio of tipped EO : random EO is 3:1 to 1:3.

Such prepolymers are made in a conventional way by allowing the polyisocyanate and polyol to react at ambient or elevated temperature (e.g. up to 90°C) and if desired in the presence of a catalyst. The MDI may be pure 4,4'-MDI or a mixture with 2,4'-MDI optionally together with minor amounts (less than 5% by weight) of 2,2'-MDI. The polyol used to make the prepolymer is also known in the art; see EP 609982. Preferably the average molecular weight is 3000-9000 and the average nominal hydroxy functionality is 2-4.

The polyisocyanate composition according to the present invention is a composition of 1-99, preferably 5-95 and most preferably 10-90 parts by weight (pbw) of the prepolymer according to the present invention with 1-99, preferably 5-95 and most preferably 10-90 pbw of another polyisocyanate, calculated on 100 pbw of this polyisocyanate composition. The other polyisocyanate may be selected from aliphatic, cycloaliphatic and araliphatic polyisocyanates, especially diisocyanates, like hexamethylene diisocyanate, isophorone diisocyanate, cyclohexane-1,4-diisocyanate, 4,4'-dicyclohexylmethane diisocyanate and m-and p-tetramethylxylylene diisocyanate, and in particular aromatic polyisocyanates like tolylene diisocyanates (TDI), phenylene diisocyanates and most preferably diphenylmethane diisocyanates (MDI) having an isocyanate functionality of at least two. The diphenylmethane diisocyanates (MDI) may be selected from pure 4,4'-MDI, isomeric mixtures of 4,4'-MDI and 2,4'-MDI and less than 5% by weight of 2,2'-MDI, crude and polymeric MDI having isocyanate funtionalities above 2, and modified variants of these MDIs containing urethane groups, obtained by reacting such MDIs with polyols having a molecular weight of at most 500, and/or carbodiimide and/or uretonimine groups, such variants having an NCO-value of at least 20% by weight and mixtures of such diphenylmethane diisocyanates having an isocyanate functionality of at least 2.

Examples of MDI, uretonimine/carbodiimide modified MDI and polymeric MDI are Suprasec MPR, 2020 and 2185 respectively, all ex Huntsman Polyurethanes, (Suprasec is a trademark of Huntsman International LLC). The variants containing urethane groups are also widely known and commercially available. The compositions according to the present invention may be made by simple mixing, high shear mixing or in-line blending or any other way of mixing the ingredients.

In particular prepolymers having a lower NCO-value, e.g. 5-19% by weight, may be mixed with such further polyisocyanates so as to prepare a prepolymer having a higher NCO-value which is then used to prepare the foam.

The prepolymer and the composition according to the present invention are useful for making flexible polyurethane foams. Therefore the present invention is further concerned with a process for preparing a flexible polyurethane foam by reacting in a mould and at an index of 70-120 and preferably of 80-105:

a) a prepolymer or composition according to the present invention

b1) 30-100 parts by weight of a polyoxyethylene-polyoxypropylene polyol, having an average nominal hydroxy functionality of 2-6 and an oxyethylene (EO) content of more than 50% by weight;

b2) 0-70 parts by weight of a polyoxyethylene-polyoxypropylene polyol, having an average nominal hydroxy functionality of 2-6, an EO content of between 20-50% by weight and a primary hydroxy content of at least 50%, calculated on the number of primary and secondary hydroxyl groups in polyol b2), the amounts of b1) and b2) being calculated on 100 parts by weight of b1) and b2); and

c) water; and optionally

d) additives and auxiliaries known per se.

In the context of the present invention the following terms, if and whether they are used, have the following meaning :

1) isocyanate index or NCO index or index :

the ratio of NCO-groups over isocyanate-reactive hydrogen atoms present in a formulation, given as a percentage :

$$\frac{[\mathrm{NCO}] \times 100}{[\text{active hydrogen}]} \quad (\%).$$

In other words the NCO-index expresses the percentage of isocyanate actually used in a formulation with respect to the amount of isocyanate theoretically required for reacting with the amount of isocyanate-reactive hydrogen used in a formulation.

It should be observed that the isocyanate index as used herein is considered from the point of view of the actual foaming process involving the isocyanate ingredient and the isocyanate-reactive ingredients. Any isocyanate groups consumed in a preliminary step to produce modified polyisocyanates (including such isocyanate-derivatives referred to in the art as prepolymers) or any active hydrogens reacted with isocyanate to produce modified polyols or polyamines, are not taken into account in the calculation of the isocyanate index. Only the free isocyanate groups and the free isocyanate-reactive hydrogens (including those of the water) present at the actual foaming stage are taken into account.

2) The expression "isocyanate-reactive hydrogen atoms" as used herein for the purpose of calculating the isocyanate index refers to the total of hydroxyl and amine hydrogen atoms present in the reactive compositions in the form of polyols, polyamines and/or water; this means that for the purpose of calculating the isocyanate index at the actual foaming process one hydroxyl group is considered to comprise one reactive hydrogen, one primary or secondary amine group is considered to comprise one reactive hydrogen and one water molecule is considered to comprise two active hydrogens.

3) Reaction system : a combination of components wherein the polyisocyanate component is kept in a container separate from the isocyanate-reactive components.

4) The expression "polyurethane foam" as used herein generally refers to cellular products as obtained by reacting polyisocyanates with isocyanate-reactive hydrogen containing compounds, using foaming agents, and in particular includes cellular products obtained with water as reactive foaming agent (involving a reaction of water with isocyanate groups yielding urea linkages and carbon dioxide and producing polyurea-polyurethane foams).

5) The term "average nominal hydroxyl functionality" is used herein to indicate the average functionality (number of hydroxyl groups per molecule) of a polyol on the assumption that this is the average functionality (number of active hydrogen atoms per molecule) of the initiator(s) used in their preparation although in practice the functionality of the polyol will often be somewhat less because of some terminal unsaturation. The average equivalent weight of a polyol is the average molecular weight divided by this average nominal hydroxyl functionality.

6) The term "average" is used to indicate an average by number.

The following way of describing polyols is used in the present application : A PO-EO polyol is a polyol having first a PO block attached to the initiator followed by an EO block (PO stands for oxypropylene and EO for oxyethylene). A PO-PO/EO polyol is a polyol having first a PO block and then a block of randomly distributed PO and EO. A PO-PO/EO-EO polyol is a polyol having first a PO block then a block of randomly distributed PO and EO and then a block of EO. A PO-EO polyol is a polyol having first a PO block and then an EO block. In the above descriptions only one tail of a polyol is described (seen from the initiator); the nominal hydroxy functionality will determine how many of such tails will be present.

Polyol b1 is an EO rich polyol. It can be prepared by known methods. It comprises PO and EO, where the EO can be random, tipped, or both. Preferably the EO is random in majority. The EO content is greater than 50% by weight and preferably 60-90% by weight calculated on the weight of the polyol. Preferably the average nominal hydroxy functionality is 2-4. The average molecular weight may range from 2000-10000, preferably from 3000-8000. Examples of useful polyols are Daltocel F442, F444 and F555, all ex-Huntsman Polyurethanes. Daltocel is a trademark of Huntsman International LLC.

Polyol b2, when used, preferably has a structure of the type PO-PO/EO-EO, PO/EO-EO or PO-EO. The total EO content is between 20-50% by weight and preferably from 21 to 45% calculated on the weight of the polyol. Polyol b2 has a primary OH content of at least 50%, preferably at least 70%. In the PO-PO/EO-EO type polyol -which is the most preferred one- the (first) PO block comprises preferably from 60 to 90% by weight of the PO and the weight ratio of tipped EO and random EO preferably is from 1:3 to 3:1. The polyol having a structure of the type PO-PO/EO-EO can notably be produced according to the teaching of Chaffanjon et al. US-A-5594097. The polyol having a structure of the type -PO/EO-EO can notably be produced according to the teaching of the Hostettler US-A-4559366. The average nominal hydroxy functionality preferably is 2-4. The average molecular weight may range from 2000-10000 and preferably from 3000-9000.

Preferably no polyols are used which have an average molecular weight of 2000 or more and an oxyethylene content of less than 20% by weight.

Water is used as the blowing agent. Carbon dioxide may be added if needed. It is appropriate to use from 1 to 10% preferably from 1 to 5 % by weight of water based on the weight of the total polyol component (prereacted and not prereacted, i.e. the total starting polyol compounds).

Other conventional ingredients (additives and/or auxiliaries) may be used in making the polyurethanes. These include catalysts, for example, tertiary amines and organic tin compounds, surfactants, cross linking or chain extending agents, for example, low molecular weight compounds such as diols, triols and diamines, flame proofing agents, for example, halogenated alkyl phosphates, fillers and pigments. Foam stabilizers, for example polysiloxane-polyalkylene oxide block copolymers, may be used to stabilize or regulate the cells of the foam. The amount of other isocyanate-reactive ingredients other than polyols b1) and b2) and water preferably is less than 15% and more preferably less than 10% by weight calculated on the weight of polyol b1) + b2).
The amount of these minor ingredients used will depend on the nature of the product required and may be varied within limits well known to a polyurethane foam technologist.

The components of the polyurethane forming reaction mixture may be mixed together in any convenient manner. They may be pre-blended so as to reduce the number of component streams to be brought together in the final mixing step. It is often convenient to have a two-stream system whereby one stream comprises the prepolymer or polyisocyanate composition and the second stream comprises all the other components of the reaction mixture. All the other components may also be led to the mixing head of the foaming machine independently.

The foams may be used in the furniture and automotive industries in seating, steering wheels, dash boards, cushioning and mattresses, and for sound damping and insulation.

The foams thus obtained have an overall density of 15-140 kg/m^3, preferably 30-110 kg/m^3 (ISO 845).

Foams made from a prepolymer or composition according to the present invention show a better stability than foams made according to the one-shot process and show a higher hardness than foams made from prepolymers based on polyol b1.

The process may be conducted in any type of mould known in the art. Preferably the process is conducted in a closed mould. Examples of such moulds are the moulds commercially used for making polyurethane furniture parts, automotive seating and other automotive parts, like arm-rests and head-rests. The moulding process is a so-called cold-cure moulding process wherein the ingredients used for making the foam are fed into the mould at a temperature of from ambient temperature up to 80°C and preferably up to 55°C, the mould being kept at a temperature of from ambient temperature up to 80°C and preferably up to 55°C during the process. After demoulding the foams are optionally cured for a period of 1 hour to 2 days at a temperature of ambient to 100°C and preferably of ambient temperature to 70°C.

The process according to the present invention may be conducted as a so-called foam-in-fabric (FIF) process (or also called pour-in-place process).

According to this process the ingredients to make the foam are poured into a mold containing an upholstery material or outer layer, such as rayon, vinyl, treated natural fibres and the like. A major problem with this approach is that the reacting liquid urethane mixture can strike through or penetrate the upholstery material while the latter is conforming to the shape of the mold, and thus to the shape of the desired composite article.

"Strike-through" is a problem because the reacting polyurethane mixture is poured inside the upholstery where the foaming action moves it, under pressure, toward the walls of the mould. Because of the pressure exerted on the upholstery, the urethane mixture is prone to penetrate the upholstery material and result in a defective article. Often a thin foam barrier is used in place between the foam and the upholstery. However, penetration of the foam barrier is also not preferred since even though such strike-through is not visible, there is a harsh feeling to the touch at the point of penetration.

Also often an impermeable film is used between the upholstery material and the thin foam barrier. By using the ingredients according to the present invention in this pour-in-place technique no such film is needed and no or less strike-through is observable. Therefore the process according to the present invention advantageously is conducted in a mould comprising an upholstery material.

Conventional polyurethane systems which use primarily polyether polyols comprising predominantly oxypropylene groups normally need a barrier film certainly at higher overpack; surprisingly the present systems can do without barrier films. The overpack in the present foam in fabric process preferably is 5-100 and more preferably 10-50%. In particular the use of the prepolymer based on the polyol of the type PO-PO/EO-EO allows a higher overpack without observing too severe penetration when using the foam - in-fabric technique.

Since the foams according to the present invention can be easily demoulded without applying external mould release after the first part has been made, the process for preparing the flexible polyurethane foam in a closed mould may be conducted according to the following steps :

1. an external mould release agent is applied onto at least those surfaces of the mould which will be in contact with the ingredients used for preparing the foam and/or the finished foam;
2. the ingredients to be used for preparing the foam are fed into the mould;
3. the ingredients are allowed to react and to form the polyurethane foam which reaction comprises reacting the ingredients described before;
4. the polyurethane foam so formed is removed from the mould and
5. steps 2,3 and 4 are repeated at least 10 times without repeating step 1.

The foams obtained have comparable physical properties irrespective of whether the foam is obtained after steps 2,3 and 4 have been conducted once, 10 times, 25 times, 40 times or even more.

The mould may be closed after step 2, and opened after step 3 or the mould may be closed after step 1 and opened after step 3; in the latter case the ingredients for making the foam are fed into the mould via appropriate inlets.

The material of the mould may be selected from those known in the art like metal, e.g. steel, aluminium, and epoxy resin.

Step 1 of the process may be conducted in any way known in the art. Applying an external mould release agent on the surfaces of a mould, which surfaces will be in contact with the ingredients used for making the material and/or with the material includes any way of applying such an agent to the surfaces, like rubbing, brushing, spraying and combinations thereof and applying any agent or agents intended to facilitate the later demoulding. One or more external mould release agents may be used as well as mixtures of external release agents.

The external mould release agents may be applied as such or as a solution, emulsion or dispersion in a liquid.

The external mould release agents, applied in step 1, may be applied in one or more stages. Any external mould release agent known in the art may be applied; examples of suitable external mould release agents are Kluberpur 41-0039 and 41-0061 (both from Kluber Chemie), Desmotrol D-10RT from Productos Concentrol SA, Acmosil 180 STBH from Fuller and Johnson Cire 103 from Johnson and Johnson.

The invention is illustrated by the following examples :

Example 1

72 parts by weight (pbw) of polyol 1 was heated to about 45°C and added to a mixture of 28 pbw of Suprasec MPR and about 10 ppm of thionylchloride which mixture was kept at about 50°C. Then the reaction was allowed to proceed while keeping the temperature at 85°C for about 3½ hours.

Subsequently, the prepolymer obtained was allowed to cool to ambient temperature. The prepolymer (hereinafter "Prepolymer 1") had an NCO-value of about 7.8% by weight.

Polyol 1 is a polyol with the structure PO-PO/EO-EO which is glycerol-initiated and has an OH-value of 28 mg KOH/g and a weight distribution of about 55-15/15-15 respectively.

Example 2

Polyisocyanate compositions according to the present invention were made by mixing the following polyisocyanates (amounts are in pbw).

Polyisocyanate composition	1	2	3	4	5
Prepolymer 1	36.3	20.9	50.6	27.2	48.4
Suprasec 2020	12.1	11.1	4.8	30.8	20.8
Suprasec MPR	8.9	11.1	7.6	30.8	20.8
Suprasec 2185	3.4	1.1	1.2	11.1	10.0

Example 3

Moulded flexible foams were made from the compositions of example 2 and the following further ingredients which were premixed before being brought in contact with the polyisocyanates.

The mould temperature was 45-50°C, the temperature of the chemicals before mixing was 27°C. After feeding the mixture into the mould, the mould was closed. The mould size was 45x45x10 cm and the amount of overpack was about 50%.

The ingredients and the physical properties of the moulded foams obtained are given in below table.

Foam	1	2	3	4	5
Polyisocyanate composition	1	2	3	4	5
Polyol 2	-	-	33.6	62.9	95
Polyol 3	36.5	43.5	-	-	-
Polyol 1	-	10	-	32.1	-
ZF22	0.06	0.04	0.06	-	-
Dabco 33 LV	0.45	0.4	0.4	-	-
B8715 LF	0.25	0.15	0.15	0.45	0.5
water	2.04	1.64	1.54	3.60	3.43

D8154	-	-	-	0.53	0.6
DMEA	-	-	-	0.27	0.3
DMAPA	-	-	-	0.16	0.2
core density, kg/m^3, ISO 845	57	69	68	59	-
transmissibility at resonance, JASO B407-82	3.1	3.0	3.2	-	-
Indentation load deflection, N/$_{314}$cm^2, at 25%, ISO 2439	265	246	236	-	-
Creep (compression load deflection 40%), kPa	8.5	-	9.8	8.3	-

Polyol 2 : is a polyoxyethylene polyoxypropylene polyol having a nominal functionality of 3, an OH-value of 38 mg KOH/g and an EO content of about 75% by weight (all random).

Polyol 3 : as Polyol 2 with the proviso that the OH-value is 28 mg KOH/g.

ZF22 : Jeffcat ™ ZF22, a catalyst obtainable ex Huntsman.

Dabco 33 LV : catalyst ex Air Products

B8715LF : Tegostab 8715 LF a surfactant from Goldschmidt

D8154 : Blend of tert. amine catalyst and organic acid catalyst ex Air Products.

DMEA : dimethyl ethanolamine catalyst.

DMAPA : dimethylamino-propylamine catalyst.

CLAIMS

1. Isocyanate-terminated prepolymer having an NCO-value of 5-30% by weight and being the reaction product of an excessive amount of diphenylmethane diisocyanate comprising at least 80% by weight of 4,4'-diphenylmethane diisocyanate and a polyoxyethylene-polyoxypropylene polyol having an average molecular weight of 2000-10000, an average nominal hydroxy functionality of 2-6, an oxyethylene content of 21-45% by weight and a structure of the type -PO-PO/EO-EO wherein the PO block comprises 60-90% of the PO and the ratio of tipped EO : random EO is 3:1 to 1:3.

2. Prepolymer according to claim 1 wherein the diisocyanate comprises at least 90% by weight of 4,4'-diphenylmethane diisocyanate and the polyol has an average molecular weight of 3000-9000 and the average nominal hydroxy functionality is 2-4.

3. Prepolymer according to claims 1-2 wherein the NCO-value is 5-19% by weight.

4. Polyisocyanate composition comprising 1-99 parts by weight of a prepolymer according to claims 1-3 and 1-99 parts by weight of another polyisocyanate, calculated on 100 parts by weight of this polyisocyanate composition.

5. Composition according to claim 4 wherein the amounts are 10-90 parts by weight.

6. Composition according to claims 4-5 wherein the other polyisocyanate is selected from 1) diphenylmethane diisocyanates (MDI), 2) crude MDI, 3) polymeric MDI, 4) modified variants of these MDIs containing urethane groups, obtained by reacting these MDIs with polyols having a molecular weight of at most 500, and/or carbodiimide and/or uretonimine groups, such

variants having an NCO-value of at least 20% by weight and 5) mixtures of any of the polyisocyanates 1-4.

7. Process for preparing a flexible polyurethane foam by reacting in a mould and at an index of 70-120 :

a) a prepolymer or composition according to claims 1-6;

b1) 30-100 parts by weight of a polyoxyethylene-polyoxypropylene polyol, having an average nominal hydroxy functionality of 2-6 and an oxyethylene (EO) content of more than 50% by weight;

b2) 0-70 parts by weight of a polyoxyethylene-polyoxypropylene polyol, having an average nominal hydroxy functionality of 2-6, an EO content of between 20-50% by weight and a primary hydroxy content of at least 50%, calculated on the number of primary and secondary hydroxyl groups in polyol b2), the amounts of b1) and b2) being calculated on 100 parts by weight of b1) and b2); and

c) water; and optionally;

d) additives and auxiliaries known per se.

8. Process according to claim 7 wherein polyol b2) has the structure PO-EO/PO-EO wherein the PO block comprises 60-90% by weight of the PO and wherein the weight ratio of tipped EO and random EO is 1:3 to 3:1.

9. Process according to claims 7-8, wherein no polyol is used which has an average molecular weight of 2000 or more and an oxyethylene of less than 20% by weight.

10. Process according to claims 7-9, wherein the process is conducted in a closed mould.

INTERNATIONAL SEARCH REPORT

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PCT/EP 03/07053

A. CLASSIFICATION OF SUBJECT MATTER
IPC 7 C08G18/10 C08G18/48

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 7 C08G

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)

WPI Data, EPO-Internal, PAJ

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category °	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	US 5 792 829 A (ALLEN ET AL) 11 August 1998 (1998-08-11) column 5, line 57 -column 7, line 30 column 11, line 52 -column 12, line 7 column 14, line 29 - line 53; claims 1,6-9; example 4 ---	1
A	WO 01 32736 A (HUNTSMAN INTERNATIONAL) 10 May 2001 (2001-05-10) cited in the application page 5, line 22 -page 7, line 10; claim 1 -----	1

☐ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

° Special categories of cited documents :

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