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(54) Title: PROCESS FOR THE PRODUCTION OF AN ETHYLENE POLYMER USING A CHROMIUM CONTAINING CATALYST

(57) Abstract: The invention is directed to a process for the production of high density polyethylene by polymerisation of ethylene in the presence of a supported chromium oxide based catalyst and an activator characterised in that the activator comprises the reaction mixture of a boron compound and/or an alkyl aluminium compound and a nitrogen containing compound wherein the boron compound is a (C₁- C₁₀) alkyl boron compound or a (C₅- C₂₀) aromatic boron compound, wherein the alkyl aluminum compound is an organo aluminium compound having the formula AIR₃ in which R is a hydrocarbon radical containing 1 -10 carbon atom and wherein the nitrogen containing compound comprises -NH₂, -NHR, -NR₂, wherein R may be an alkyl or a substituted alkyl having from 1 to 40 carbon atoms.



PROCESS FOR THE PRODUCTION OF AN ETHYLENE POLYMER USING A CHROMIUM CONTAINING CATALYST

The invention relates to the polymerisation of high density ethylene in the presence of a supported chromium oxide based catalyst.

The production processes of LDPE, HDPE and LLDPE are summarised in "Handbook of Polyethylene" by Andrew Peacock (2000; Dekker; ISBN 0824795466) at pages 43-66. The catalysts can be divided in three different subclasses including Ziegler Natta catalysts, Phillips catalysts and single site catalysts. The various processes may be divided into solution polymerisation processes employing homogeneous (soluble) catalysts and processes employing supported (heterogeneous) catalysts. The latter processes include both slurry and gas phase processes.

The polymerisation of ethylene with supported chromium based catalysts is disclosed by Kevin Cann in "Comparison of silyl chromate and chromium oxide based olefin polymerisation catalysts" (Macromolecular Symp, 2004, 213, 29-36).

The chromium oxide based catalyst, which is commonly referred to in the literature as "the Phillips catalyst", can be obtained by calcining a chromium compound carried on an inorganic oxide carrier in a non-reducing atmosphere. The chromium oxide catalysis and the ethylene polymerisation with this specific catalyst are disclosed in "Handbook of Polyethylene" by Andrew Peacock at pages 61-64.

The silyl chromate catalyst consists of silylchromate (bis-triphenylsilyl chromate) absorbed on dehydrated silica and subsequently reduced with for example diethylaluminum ethoxide. The silylchromate catalysts are more expensive than chromium oxide catalysts.

Although similar in structure the oxo and triphenylsilyl chromate catalyst perform very differently in polymerisation reactions. Silylchromate-based catalysts generally produce desirable polyethylenes relative to those produced by chromium oxide-type catalysts. Silylchromate produced polyethylenes generally have a broader molecular weight distribution than those produced using chromium oxide-type catalysts. The broader molecular weight distribution leads to better processability of the

resulting polyethylene. However, the productivities of silylchromate-based catalysts are typically much poorer than those realized using chromium oxide-based catalysts.

Chromium oxide-on-silica catalysts result in good
5 production rates however certain applications require a broader molecular weight distribution.

Another disadvantage of the polymers produced with silylchromate-based catalysts is that benzene may be present in the end product which limits its application in food packaging. It is highly desirable
10 to have a benzene free catalyst system in combination with the high productivity of the chromium oxide-on-silica catalysts and at the same time is able to produce high density polyethylene (HDPE) with broad or bimodal molecular weight distributions.

It is the object of the present invention to provide a high
15 density ethylene polymerisation process in the presence of chromium oxide based catalysts which results in high density ethylene polymers with a broader molecular weight distribution (MWD) while maintaining the other required characteristics for example high catalyst activity and polymer yield.

The process according to the invention is characterised in
20 that high density ethylene polymer is obtained by polymerizing ethylene in the presence of a supported chromium oxide based catalyst and an activator comprising a reaction mixture of a boron compound and/or an alkyl aluminum compound and a nitrogen containing compound.

The combination of the supported chromium oxide based
25 catalyst and the specific activator comprising the reaction product of the boron compound and/or the alkyl aluminum compound and a nitrogen containing compound results in a broader MWD of the polyethylene.

Furthermore the combination of the chromium catalyst and the activator results in a high productivity of the high density ethylene
30 polymerisation process.

Another advantage of the process according to the present invention is that the obtained products show a behaviour similar to the

products obtained with the silylchromate on silica catalyst.

A further advantage is the use of a benzene free catalyst system.

Furthermore bimodal HDPE polymers with broad MWD can
5 be produced with the chromium based catalyst and the activator comprising the reaction mixture of an alkyl boron compound and/or an alkyl aluminium compound and a nitrogen containing compound.

The chromium containing catalyst contains a support.
Preferably the support is a silica support. The silica may have a surface
10 area (SA) larger than $150 \text{ m}^2/\text{g}$ and a pore volume (PV) larger than $0.8 \text{ cm}^3/\text{g}$. The support may be modified so as to include cogels such as for example silica-titania or silica-alumina and by the replacement of silica by alumina or amorphous aluminium phosphates. Furthermore, the support may comprise a tergel which is produced by mixing a chromium source
15 with the silica and titania compound. The chromium containing catalyst may also be doped with chemical compounds containing for example aluminium, titanium, phosphorus, boron or fluor for example by impregnation of the porous chromium containing supports with a solution of any one of these compounds.

20 Preferably, the catalyst is an unmodified silica supported chromium based catalyst having a pore volume larger than $0.8 \text{ cm}^3/\text{g}$ and a specific surface area of at least $150 \text{ m}^2/\text{g}$.

The properties of the catalyst, pore volume and specific
surface area are determined before the catalyst is activated at an elevated
25 temperature.

The amount of chromium in the catalyst is generally at least 0.5 % by weight. Preferably the amount of chromium in the catalyst is at least 1.0 % by weight.

The average particle size (D_{50}) of the catalyst may range
30 between for example 15 and 150 micrometers. Generally, the catalyst is activated before being applied in the polymerisation reaction. The activation may take place under different conditions. The activation

generally takes place at an elevated temperature, for example, at a temperature above 450°C. The activation may take place in different atmospheres, for example in dry air. Generally, the activation takes place at least partially under an inert atmosphere preferably consisting of nitrogen.

- 5 At the same time the temperature is raised slowly. It has been found to be advantageous to change from the nitrogen atmosphere to an atmosphere of dry air at a temperature of at most 700°C. The activation time after reaching the maximum temperature may last for several minutes to several hours. This activation time is at least 1 hour but it may be advantageous to
10 activate much longer.

Preferably the boron compound is a (C₁- C₁₀) alkyl boron compound or a (C₅- C₂₀) aromatic boron compound.

Suitable (C₁- C₁₀) alkyl boron compounds include for example trimethyl boron, triethyl boron and tripropyl boron.

- 15 Preferably, the alkyl boron compound is triethyl boron.

If triethyl boron is used as a promoter, generally the boron concentration in the polymerisation reactor is less than 30 ppm of boron based on the diluent. Preferably, the concentration of boron is less than 20 ppm and more preferably the concentration is less than 5 ppm boron.

- 20 The alkylaluminum compound may be selected from an organo aluminium compound having the formula AlR₃ in which R is a hydrocarbon radical containing 1 – 10 carbon atom. Suitable examples of organo aluminum compound of the formula AlR₃ include for example trimethyl aluminium, triethyl aluminium, triisobutyl aluminium, tri-n-hexyl
25 aluminium and tri octyl aluminium. Preferably trimethyl aluminium, triethyl aluminium or triisobutyl aluminium is applied.

- Examples of suitable nitrogen containing compound include nitrogen containing groups such as for example -NH₂, -NHR, -NR₂, wherein R may be an alkyl or a substituted alkyl having from 1 to
30 40 carbon atoms ,preferably from 1 to 20 carbon atoms. The nitrogen containing compound includes substituted derivatives thereof.

According to a preferred embodiment of the invention the

nitrogen containing compound is a compound having the formula $R-NH_2$ in which R represents an alkyl having from 2 to 20 carbon atoms.

According to a further preferred embodiment of the invention the nitrogen containing compounds is decylamine, undecylamine, 5 octylamine, octadecylamine, dodecylamine, N-methyloctadecylamine, N-ethyl-dodecylamine, hexadecylamine and/or N-N" dimethyl-n-octadecylamine

More preferably the nitrogen containing compound is octadecylamine, dodecylamine or hexadecylamine.

10 The process according to the present invention does not comprise pyrrole-containing compounds such as for example hydrogen pyrrolide or pyrrole, derivatives of hydrogen pyrrolide and metal pyrrolide complexes because these compounds are unsuitable to be applied as the nitrogen containing compound.

15 Generally, the molar ratio of aluminium to nitrogen ranges between 0.1: 1 and 4:1.

Generally, the molar ratio of borium to nitrogen ranges between 0.1: 1 and 4:1.

20 Generally, the molar ratio of aluminium to chromium ranges between 0.01: 100 and 100:1.

Preferably, the ratio of weight equivalents of aluminium to weight equivalents of chromium ranges between 0.1: 100 and 100:1.

The productivity of the catalyst ranges between 100 gram PE/ gram Cr-cat per hour and 50000 gram PE/ gram Cr-cat per hour by 25 selecting the amount of the chromium containing catalyst.

The polymerisation takes place in a single reactor.

The polymerisation may be performed via a gas phase process or via a slurry process.

30 Gas fluidized bed polymerisation processes are summarised by Than Chee Mun in Hydrocarbons 2003 "Production of polyethylene using gas fluidised bed reactor". Gas phase polymerisation generally involves adding gaseous monomers into a vertically oriented

polymerisation reactor filled with previously formed polymer, catalyst particles and additives. Generally the polymerisation in the gas phase polymerisation systems takes place at temperatures between 30°C and 130°C with super atmospheric pressures. The rising gas phase fluidizes the bed, and the monomers contained in the gas phase polymerize onto supported catalyst or preformed polymer during this process. Upon reaching the top of the reactor, unreacted monomer is recycled, while polymer continually falls down along the sides of the reactor. Examples of suitable gas phase polymerisations are disclosed in for example US-A- 4003712 and US-A- 2005/0137364.

In slurry reactors, a low boiling hydrocarbon solvent such as isobutane, hexane or heptane is employed as a continuous medium, and monomer, catalyst etc added to this continuous phase. The polymer formed is insoluble in the reaction medium, producing slurry of polymer and catalyst. Slurry reactors may be divided into loop reactors and boiling solvent reactors. Heat is at least partially removed by the heat of vaporization of solvent, which is later condensed and returned to the reactor. Polymer is removed as slurry from the bottom of the reactor and flashed to remove solvent, which is recycled. Slurry loop reactors may be horizontally or vertically oriented. Water flowing between the tubes serves to remove heat and maintain a relatively constant temperature. Slurry flow is achieved by pumps which maintain the polymer slurry at relatively high velocity. Product is removed either continuously or discontinuously from a "settling leg." Preferably, the polymerisation of ethylene takes place in a diluent at a temperature of between 80°C and 110°C. Suitable diluents include, for example, isobutane and propane.

In the case of the production of an ethylene copolymer the alpha olefin co monomer may be propylene, 1-butene, 1-pentene, 4-methyl-1-pentene, 1-hexene and/or 1-octene.

The activator may be added to the catalyst directly or separately. Separated injections to the reactor are preferred.

The HDPE reactor powder obtained with the process

according to the present invention has:

- a high-load melt index (HLMI) ≥ 0.1 g/10 min and ≤ 50 g/10 min (according to ISO 1133)
- 5 • $M_w / M_n \geq 10$ and ≤ 100 (according to size exclusion chromatography (SEC) measurement)
- a density ≥ 945 kg/m³ and ≤ 965 kg/m³ (according to ISO1183).

The ethylene polymers obtained with the process according to the invention may be combined with additives such as for
10 example lubricants, fillers, stabilizers, antioxidants, compatibilizers and pigments. The additives used to stabilize the copolymers may be, for example, additive packages including hindered phenols, phosphites, UV stabilisers, antistatics and stearates.

An anti static agent can be used to suppress fouling of the
15 reactor wall. Suitable anti static agents are disclosed for instance in US4182810 and EP107127.

The ethylene polymers may be extruded or blow-moulded into articles such as for example bottles, containers, fuel tanks and drums, and may be extruded or blown into films.

20 US5331070 A discloses the polymerisation of olefins in the presence of a catalyst system composition comprising chromium supported on an inorganic oxide support, a pyrrole-containing compound and a metal alkyl. The pyrrole-containing compound can be any pyrrole-containing compound such as for example hydrogen pyrrolide or pyrrole, derivatives of
25 hydrogen pyrrolide and metal pyrrolide complexes. In contrast to the process according to US5331070 A the process according to the present invention does not apply a pyrrole-containing compound.

The process according to US5331070 A produces an olefin comonomer in-situ. The obtained polymer has a decreased density and an
30 increased branching. According to Table I the polymer yield and the activity decrease because of the presence of the pyrrolide whereas the specific activator combination according to the present invention results in an

improved productivity.

DE19607888 A1 discloses a process for the oligomerization of alpha -olefins in a solvent using a chromium-based catalyst system comprising at least (a) an unsupported chromium
5 compound, (b) at least one nitrogen-containing compound selected from amines, amides and imides, and (c) an alkylaluminum compound, and the catalyst components and by-product polymers are recovered simultaneously from the reaction solution. Preferred examples of the chromium compounds are chromium alkoxides, chromium carboxylates,
10 chromium diketonates and salts of chromium with anions of ketoester and chromium halides. DE19607888 A1 does not disclose a supported chromium oxide based catalyst. The nitrogen-containing compound is selected from nitriles, amines and amides such as for example acetonitrile, pyridine, dimethylpyridine, dimethylformamide, N-methylformamide, aniline,
15 nitrobenzene, tetramethylethylenediamine, diethylamine, isopropylamine, hexamethyl disilazane and pyrrolidone.

It is the object of the process according to DE19607888 A1 to provide an industrially advantageous process for preparing alpha.-olefin oligomers which process is simple and capable of efficiently separating the
20 by-product polymers and catalyst components. In contrast to the polymerisation process according to the present invention the process according to DE19607888 A1 is directed to an oligomerisation process wherein the polymer is an undesired by-product. DE 19607888 A1 does not disclose a process for the production of high density polyethylene by
25 polymerisation of ethylene in the presence of a supported chromium oxide based catalyst and an activator comprising the reaction mixture of a boron compound and/or an alkyl aluminium compound and a nitrogen containing compound.

EP 67607 A2 discloses a catalyst containing a solid
30 catalyst component and an organometallic compound component. The solid catalyst is obtained by calcining a reaction product of (a) at least one compound selected from an amine compound, a phosphoric acid amide, a

hydro carbyloxy compound of titanium, vanadium, hafnium or zirconium and a specific germanium compound with (b) at least one chromium compound, said reaction product being supported on an inorganic oxide carrier.

5 It is the object of EP 67607 A2 to provide a catalyst suitable for use in the polymerization of olefins, which catalyst has a high activity and is capable of producing olefin polymers having excellent melt flowability. The organometallic compound component is selected from a specific hydrocarbon-soluble organomagnesium component , an
10 organoaluminum component obtained by reacting a trialkylaluminum compound with a carbinol , a silanol or siloxane, a component comprising the specific hydrocarbon-soluble organomagnesium component and an organoaluminum component , a component comprising the specific hydrocarbon-soluble organomagnesium component and an organoboron,
15 organozinc, or organolithium compound and a component comprising an organoaluminum component and an organoboron, organozinc or organolithium compound.

 The process according to EP 67607 A2 and the process according to the present invention are completely different because
20 amongst others in the case that an amine is applied in the process according to EP 67607 A2 this amine is calcined with a chromium compound. Furthermore the organoaluminum component is obtained by reacting a trialkylaluminum compound with a carbinol, a silanol or siloxane. The process according to the present invention does not comprise the
25 specific organometallic compound.

 The invention will be elucidated by means of the following non-limiting examples.

Examples

30 The properties of the polymers produced in the Examples were determined as follows:

Flow Index:

Melt Index (2.16): ASTM D-2338 Condition E measured at 190 °C reported as grams per 10 minutes.

Flow Index: (FI₂₁) ASTM D-1238 Condition F measured at 190°C using 10 times the weight as used in Melt Index above.

MFR: melt flow ratio is the flow index/melt index

Density:

The density (g/cm³) was determined as specified in ASTM D 1505-68 with the exception that the density measurement was taken 4 hours instead of 24 hours after the sample was placed in the density column.

Gel Permeation Chromatography (GPC):

Polymer molecular weight and its distribution (MWD) were determined by Polymer Labs 220 gel permeation chromatograph. The chromatograms were run at 150 °C using 1,2,4-trichlorobenzene as the solvent with a flow rate of 0.9 ml / min. The refractive index detector is used to collect the signal for molecular weights. The software used is Cirrus from PolyLab for molecular weights from GPC. The calibration of the HT-GPC uses a Hamielec type calibration with broad standard and fresh calibration with each sample set.

Experiments A-F

Preparation of an activator comprising the reaction mixture of an alkyl boron compound and/or an alkyl aluminum compound and a nitrogen containing compound

Under a dry nitrogen atmosphere, a Schlenk flask was charged with triisobutyl aluminium (TIBAL) (heptane solution), triethyl aluminium (TEAL) or triethyl boron (TEB) and a hexane solution (temperature 65 °C) of octadecylamine (C₁₈H₃₇NH₂). The molar ratio Al/N or B/N is indicated in Table 1. The compounds were mixed to form a colour solution.

Table1

	Aluminum or boron alkyl compound	Nitrogen compound	Al/N or B/N molar ratio
A	TIBAL (1M)	C ₁₈ H ₃₇ NH ₂	1
B	TIBAL (1M)	C ₁₈ H ₃₇ NH ₂	1.5
C	TIBAL (1M)	C ₁₈ H ₃₇ NH ₂	2
D	TMAL (2M)	C ₁₈ H ₃₇ NH ₂	2
E	TEAL (1M)	C ₁₈ H ₃₇ NH ₂	2
F	TEB (1M)	C ₁₈ H ₃₇ NH ₂	2

EXAMPLES I-IV AND COMPARATIVE EXAMPLE APolymerisation in slurry process

- 5 The CrO₃ catalyst (F3; Dow) was used as the catalyst in the polymerisation of ethylene and 1-butene as the comonomer. The polymerisations were carried out in a two-liter stirred autoclave, in deoxygenated isopentane. Polymerisations were carried out at 100 °C and 15 bars (15.10⁵ MPa) of total pressure. The activators according to
- 10 Experiments A-D were used in an amount of 1 ml. Ethylene gas was used to maintain this pressure. Upon completion of the polymerisation, the reactor was vented and cooled to ambient temperature to recover the polymer. Details of each polymerisation and characteristics of the resins produced are provided in Table 2.

15

Table 2

	Activator						
		Prod. (g PE/g cat.h)	MI ₅	FI	MFR	Mw	Mw/Mn
I	C	402	0.4	10.5	26	207,500	36.8
II	A	230	0.12	7.0	58	500,000	88.9
III	B	296	0.1	6.7	67	681,000	122.7
IV	D	210	0.27	11.8	44	403,000	49.7
A	TEAL	278	5.8	78.0	13	123,200	16.7

EXAMPLE V AND COMPARATIVE EXAMPLES B-CPolymerisation in a gas phase process

The polymerisations were carried out in a two-liter autoclave equipped with a helical stirrer with a CrO₃ catalyst (F3; Dow).

- 5 NaCl (300g), which has been baked for 24 hours over 120 °C was used as seed bed. Mixtures B and C were applied as activator. Polymerisations were carried out at 102°C and 20 bar (20.10⁵ MPa) of total pressure. Ethylene gas was used to maintain this pressure. Upon completion of the polymerisation, the reactor was vented and cooled to ambient temperature
- 10 to recover the mixture of NaCl and polyethylene polymer. The mixture was stirred in water for 24 hours before being filtered and dried to recover the polymer.

Table 3

	Activator	Al/Cr Molar ratio	Prod. (g PE/g cat.h)
V	B	6.7	408
B	TIBAL	6.7	167
C	TEAL	6.7	133

15

These examples show that the mixture comprising TIBAL and C₁₈H₃₇NH₂ improves the productivity.

EXAMPLES VI-VIII AND COMPARATIVE EXAMPLES D-F20 Polymerisation in a gas phase process

The polymerisations were carried out in another two-liter autoclave equipped with a helical stirrer with a CrO₃ catalyst (F3; Dow). NaCl (300g), which has been baked for 24 hours over 120 °C was used as seed bed. Mixture C was applied as activator. Polymerisations were

25 carried out at 102°C and 20 bar (20.10⁵ MPa) of total pressure. Ethylene gas was used to maintain this pressure. Upon completion of the

polymerisation, the reactor was vented and cooled to ambient temperature to recover the mixture of NaCl and polyethylene polymer. The mixture was stirred in water for 24 hours before being filtered and dried to recover the polymer.

5

Table 4

	Activator	Al/Cr	Prod. (g PE/g cat.h)
VI	C	6.7	133
VII	C	13	260
VIII	C	20	307
D	TEAL	6.7	67
E	TIBAL	6.7	93
F	TIBAL	20	87

These examples show that activator C improves the productivity.

CLAIMS

1. A process for the production of high density polyethylene by polymerisation of ethylene in the presence of a supported chromium oxide based catalyst and an activator characterised in that the activator
5 comprises the reaction mixture of a boron compound and/or an alkyl aluminium compound and a nitrogen containing compound wherein the boron compound is a (C₁- C₁₀) alkyl boron compound or a (C₅- C₂₀) aromatic boron compound, wherein the alkyl aluminum compound is an organo aluminium compound having the formula AlR₃ in which R is a
10 hydrocarbon radical containing 1 –10 carbon atom and wherein the nitrogen containing compound comprises -NH₂, -NHR, -NR₂, wherein R may be an alkyl or a substituted alkyl having from 1 to 40 carbon atoms.
2. A process according to Claim 1 characterised in that the alkyl boron compound is trimethyl boron, triethyl boron or tripropyl boron.
- 15 3. A process according to any one of Claims 1-2 characterised in that the nitrogen containing compound is a compound having the formula R-NH₂ in which R represents an alkyl having from 2 to 20 carbon atoms.
4. A process according to Claim 3 characterised in that the nitrogen containing compound is decylamine, undecylamine, octylamine,
20 octadecylamine, dodecylamine, N-methyloctadecylamine, N-ethyl-dodecylamine, hexadecylamine and/or N-N" dimethyl-n-octadecylamine
5. A process according to Claim 4 characterised in that the nitrogen containing compound is octadecylamine, dodecylamine and/or hexadecylamine
- 25 6. A process according to any one of Claims 1-5 characterised in that the organo aluminum compound of the formula AlR₃ is trimethyl aluminium, triethyl aluminium, triisobutyl aluminium, tri-n-hexyl aluminium and/or tri octyl aluminium.
7. A process according to any one of Claims 1-6 characterised in that the
30 process is a gas phase process or in a slurry process.
8. A process according to Claim 7 characterised in that the process is a gas phase process.

INTERNATIONAL SEARCH REPORT

International application No

PCT/EP2010/002174

A. CLASSIFICATION OF SUBJECT MATTER
INV. C08F10/02 C08F4/69
ADD.

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)

C08F

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)

EPO-Internal, WPI Data, INSPEC

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	US 5 331 070 A (PETTIJOHN TED M [US]; REAGEN WILLIAM K [US]; MARTIN SHIRLEY J [US]) 19 July 1994 (1994-07-19) column 1, lines 5-7 column 2, lines 24-25 column 4, lines 20-37 column 6, lines 45-47 column 5, lines 15-16 examples 1-3 table 2	1-8
A	DE 196 07 888 A1 (MITSUBISHI CHEM CORP [JP]) 5 September 1996 (1996-09-05) claim 1 ----- -/--	1-8

☒ 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

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Date of the actual completion of the international search

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INTERNATIONAL SEARCH REPORT

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C(Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	EP 0 067 607 A2 (ASAHI CHEMICAL IND [JP]) 22 December 1982 (1982-12-22) page 1, lines 19-30 page 2, line 28 - page 3, line 12 page 4, lines 1-21 page 4, lines 33-35 page 13, lines 26-32 page 15, lines 14-32 example 37 -----	1-8

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Information on patent family members

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