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(54) **Title**: STABILISED POLYOLEFIN COMPOSITION

(57) **Abstract**: Stabilized polyolefin composition comprising: - A. a copolymer of ethylene and at least one α-olefin, - B. between 25 and 300 ppm by weight of α-tocopherol relative to the component.

Stabilised polyolefin composition.

The invention relates to a stabilized polyolefin composition comprising a copolymer of ethylene and at least one α-olefin and α-tocopherol.

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The stabilization of polyolefin composition is known in the art.

Suitable stabilisers known in the art are for example synthetic (poly)phenolic compounds such as tetrakis[methylene-3-(3',5')-di-t-butyl-4-hydroxyphenyl)propionate] methane; octadecyl 3,5-di-t-butyl-4-hydroxyhydrocinnamate; 1,1,3-tris(2-methyl-4-hydroxy-5-t-butylphenyl)butane; 1,3,5-trimethyl-2,4,6-tris(3,5-di-t-butyl-4-hydroxybenzyl)benzene, bis-[3,3-bis-(4’-hydroxy-3’-t-butylphenyl butanoic acid]-glycol ester; tris(3,5-di-t-butyl-4-hydroxy benzyl)isocyanurate; 1,3,5-tris(4-t-butyl-2,6-dimethyl-3-hydroxy-benzyl)isocyanurate; 5-di-t-butyl-4-hydroxy hydrocinnamic acid triester with 1,3,5-tris(2-hydroxyethyl)-s-triazine-2,4,6(1H, 3H, 5H)trione; p-cresol/dicyclopentadiene butylated reaction product and 2,6-bis(2’-bis-hydroxy-3’-t-butyl-5’-methyl-phenyl-4-methyl-phenol).

The stabilized polyolefin compounds may be processed via for example injection moulding, blow moulding, extrusion moulding, compression moulding or thin-walled injection moulding techniques. The obtained products may be applied in a huge amount of applications for example in food contact packaging applications, biomedical applications, health care applications or pharmaceutical applications.

Most of the synthetic (poly)phenolic antioxidants used are strictly regulated by governments because they are suffering from serious limitations. Synthetic antioxidants can thereby diffuse into the surrounding medium. This can in turn lead to contamination of food and/or other human-used products with potentially toxic by-product substances. This problem may arise simply because some antioxidants are toxic above a certain level of concentration.

Accordingly, the use of such antioxidants is already an issue in for example food- and water contact applications, in medical and pharmaceutical devices. With increased consumer concerns about the amount of chemicals in their foods, processors are looking for improved ways to protect their products.

There is a continuous need to provide improved stabilized polyolefin compositions having no dangerous effects when dispersed in the environment and/or which also fulfill all requirements related to processing or short term heat stabilization and/or which shows low yellowness increase during multiple extrusion.

The invention is characterized in that the stabilized polyolefin composition comprises and/or consists of:

- A. a copolymer of ethylene and at least one α-olefin,
- B. between 25 and 300 ppm by weight of α-tocopherol relative to the component A.

The use of this package results in the ability to introduce a bio-based antioxidant package with excellent resistance to oxidative degradation.

Additional advantages of the stabilizer composition according to the invention are the presence of less discoloration of the material after recycling.

Furthermore the stabilizer composition according to the invention results in improved processing stability and/or improved organoleptic properties.

The stabilized polyolefin composition is efficiently stabilized, while limiting and/or avoiding the use of toxic substances. This may especially for example allow to improve processing stability, especially by reducing cross-linking during extrusion and/or multiple extrusions. This may also for example allow to reduce yellowness increases during extrusion and/or multiple extrusions.

A further advantage is that the stabilizer composition according to the invention results in improved organoleptic properties. Moreover, the risk related to and/or the amount of compound(s) possibly migrating out of the stabilized polyolefin composition may be reduced.

Component A may be a copolymer of ethylene with at least one α-olefin, preferably a linear low density polyethylene (LLDPE). A copolymer of ethylene with at least one α-olefin may thereby preferably be a copolymer comprising ethylene and at least one α-olefin or a copolymer of ethylene and only one α-olefin.

Linear low-density polyethylene (LLDPE) may for example be obtained by polymerizing ethylene with at least one α-olefin, which may be selected from 1-butene, 1-pentene, 4-methyl-1-pentene, 1-hexene, 1-heptene and/or 1-octene, preferably 1-butene.

Linear low-density polyethylene (LLDPE) may be produced for example using at least one or exactly one metallocene catalyst or at least one or exactly one Ziegler-Natta catalyst.

Preferably, the linear low-density polyethylene (LLDPE) used according to the invention may be produced for example using at least one Ziegler-Natta catalyst comprising Mg and at least one or one of Ti, Hf or Zr. This may further contribute to an improved stabilization.

For example, the polymerized α-olefin may for example account for between 0.05 w% and 15 w%, preferably between 0.1 w% and 12 w%, further preferred between 1 w% and 10 w% , further preferred between 0.1 w% and 5 w% or between > 5 w% and < 10 w% compared to the total weight of the linear low density polyethylene.

A copolymer of ethylene and at least one α-olefin may for example have a density as determined according to ISO 1183-1 (2012), method A of ≥ 850 kg/m^3 and ≤ 950 kg/m^3, preferably ≥ 910 kg/m^3 and ≤ 940 kg/m^3, further preferred between 913 kg/m^3 and 923 kg/m^3, further preferred between > 915 kg/m^3 and < 921 or < 920 kg/m^3. This may lead to mechanical properties suitable for the use for film applications.

A copolymer of ethylene and at least one α-olefin may for example have an MFI represented by the corresponding melt mass flow rate (MFR) as measured according to ISO 1131-1 (2011) at 190°C and at a load of 2.16 kg of between 0.1 dg/min and 10 dg/min, preferably between 0.5 dg/min and 5 dg/min, further preferred between 0.5 dg/min and 3 dg/min, even further preferred >0.5 dg/min and < 2 dg/min, even further preferred >0.7 dg/min and < 1.5 dg/min. This may lead to a processability suitable for the use for film applications.

A copolymer of ethylene and at least one α-olefin, especially LLDPE, as used according to the invention may for example have an Mw/Mn between 4 and 12, preferably between 4.5 and 11, further preferred between 4.7 and 10, further preferred between 5 and 8, further preferred between > 5 and < 7, further preferred between > 5.5 and < 6.5 and/or have an Mz/Mw between 4 and 12, preferably between 4.5 and 11, further preferred between 4.7 and 10, further preferred between 5 and 8, further preferred between > 5 and < 7, further preferred between > 5.8 and < 6.8. This may further contribute to an improved stabilization.

LLDPE may preferably be produced using a gas phase or slurry process. The production processes of polyethylenes are summarised in “Handbook of Polyethylene” by Andrew Peacock (2000; Dekker; ISBN 0824795466) at pages 43-66.

Component B may be α-tocopherol. Component B may preferably be added for example as a liquid and/or using a master batch comprising between 0.5 w.-% and 5 w.-%, preferably between 1 and 5 w.-% of α-tocopherol and/or between 5 and 40 w.-%, preferably between 5 and 25 w.-%, further preferred between 5 and 15 w.-% of component C and the rest of the master batch up to 100 w.-% , for example between 80 and 94 w.-%, preferably between 75 and 95 w.-%, further preferred between 75 and 85 w.-% being a further polyolefin as component D, preferably low density polyethylene.

According to the invention the α-tocopherol may be preferably synthetic α-tocopherol.

A suitable example of α-tocopherol and/or synthetic α-tocopherol according to the invention may be is Irganox E 201 (supplied by BASF) which is a racemic mixture of equal amounts of all eight possible stereoisomers of α-tocopherol (RRR, SSS, RRS, RSR, SSR, SRS, SRR, RSS) and is referred to as dl-α-tocopherol or all-rac-alpha-tocopherol.

The use of α-tocopherol according to the invention may for example allow to achieve efficient stabilization with relatively low loadings of α-tocopherol.

The stabilized polyolefin composition may preferably not comprise other tocopherols and/or comprise no tocotrienol and/or no natural vitamin E and/or no other antioxidant and/or no other compound comprising at least one phenolic motif. The stabilized polyolefin composition may preferably comprise only α-tocopherol as antioxidant. This may allow an efficient stabilization of the stabilized polyolefin composition, while reducing the risk associated with and/or the amount of compounds that may migrate out of the stabilized polyolefin composition. This may thus reduce possible health hazards.

The amount of component B ranges between 25 ppm and 300 ppm by weight, preferably between 50 ppm and 200 ppm by weight, further preferred between > 75 ppm and < 175 ppm by weight, further preferred between > 90 ppm and 170 or <170 ppm, further preferred between > 110 ppm and < 170 ppm and/or between > 50 ppm and <100 ppm, further preferred > 60 ppm and <90 ppm by weight, relative to the component A. This may allow an efficient stabilization of the stabilized polyolefin composition, while reducing the risk associated with and/or the amount of compounds that may migrate out of the stabilized polyolefin composition. This may thus reduce possible health hazards and may further contribute to an improved stabilization.

Besides components A and B, the stabilized polyolefin composition according to the invention may also comprise component C, which may be at least one organometallic stearate such as for example magnesium stearate, aluminum stearate, sodium stearate and calcium stearate and/or at least one inorganic hydrotalcites, such as for example DHT4A , preferably calcium stearate. An organometallic stearate and/or an inorganic hydrotalcite may improve processability.

The amount of optional component C may range between 100 ppm and 1000 ppm by weight, more preferably between 200 ppm and 800 ppm by weight, further preferred between 400 ppm and 600 ppm by weight, relative to the component A.

The weight ratio B:C may range for example between 0.05:1 and 0.5:1, preferably 0.07:1 to 0.3:1, further preferred between 0.1:1 and 0.2:1.

Another advantage of the stabilized polyolefin composition may be an improved color retention after recycling.

Besides components A, B and C, the stabilized polyolefin composition according to the invention may also comprise component D, which may be at least one polyolefin, preferably a low density polyethylene. The amount of compound D may be between 0 and 10 000 ppm, preferably between 2000 and 7000 ppm.

In a stabilized polyolefin composition according to the invention, the increase of MFR between the 1st and the 6th extrusion may preferably for example be between > 0 and <0.5 g/10min, preferably between > 0 and <0.2 g/10min and/or the increase of yellowness index may preferably for example be between the 3rd and the 6th extrusion may be > 0 and <0.7, preferably > 0 and < 0.5, and/or the OIT after the 6th extrusion may preferably for example be between 2. 5 and 18 min.

A stabilized polyolefin composition according to the invention may preferably for example comprise and/or consist of:

- A. a copolymer of ethylene and at least one α-olefin, whereby the copolymer of ethylene and at least one α-olefin is linear low-density polyethylene (LLDPE) produced using at least one Ziegler-Natta catalyst, whereby further the linear low-density polyethylene (LLDPE) is produced using at least one Ziegler-Natta catalyst comprising Mg and at least one or one of Ti, Hf or Zr,

- B. between 75 and 175 ppm by weight of α-tocopherol relative to the component A and
- C. between 100 ppm and 1000 ppm by weight of a stearate, preferably calcium stearate, relative to the component A, whereby further the stabilized polyolefin composition comprises only α-tocopherol as antioxidant.

That the stabilized polyolefin composition consists of the components listed above may thereby preferably mean that no other antioxidant and/or no other stearate or hydrotalcite and/or no other compound/component is present.

The composition according to the invention may be used in the production of specific articles. Examples of preferred articles are films and/or pouches, especially for packaging applications such as food and/or beverage packaging applications, for biomedical applications for example in-vivo applications, health care applications and/or pharmaceutical applications, medical applications, especially films and/or pouches for biomedical and/or health care, and/or pharmaceutical and/or medical applications.

The present invention also concerns the use of a stabilized polyolefin composition according to the invention for films and/or pouches, especially for packaging applications such as food and/or beverage packaging applications, for biomedical applications for example in-vivo applications, health care applications and/or pharmaceutical applications, medical applications, especially films and/or pouches for biomedical and/or health care, and/or pharmaceutical and/or medical applications.

The invention will now be elucidated by way of the following examples without however being limited thereto.

Examples

The following compounds were used:

- linear low density polyethylene (LLDPE): SABIC® LLDPE 118NE (without additives (no stabilizers, no acid scavengers)
- α-tocopherol purchased from BASF (Irganox® E 201)
- calcium stearate PLC (vegetable based) supplied by Faci S.p.A.

Multiple Extrusion Experiments:

A Leistritz Micro 27/GL24 Schnecke 10 was used under air. Multiple-pass extrusion involves repeatedly passing the polymer through an extruder and then collecting the samples after each pass. After the compounding extrusion (first extrusion step), the pellets were re-extruded five more times with samples being taken after each pass through the extruder.

Melt Flow Index Measurements:

Melt stability was characterized after each extrusion step using melt flow index measurements (MFI) by measuring the melt flow rate after each extruder pass. MFI was measured by applying a standard load of 2.16 kg at a melt temperature of 190°C, in accordance with ASTM D 1238-13. Results are shown below in Fig. 1.

Oxidation Induction Time (OIT) Measurements:

Oxidation Induction Time (OIT) has been measured after each extrusion step according to ISO 11357-6: Plastics -- Differential scanning calorimetry (DSC) -- Part 6: Determination of oxidation induction time (isothermal OIT) in duplicate.

Temperature: 200°C

Heating rate: 20°C/min

Results are shown below in Fig. 2.

Yellowness Index Measurements:

Yellowness index data was obtained from instrumental color measurements with following equipment and under the following conditions:

- Spectrophotometer CM-5 (Konica Minolta)
- Light source D65
- 10° viewing angle
- Diffuse illumination (8° viewing)
- Reflectance, SCE (Specular component excluded)
- Conforms to ASTM E313 (96) standard

Results are reported in Table 2 below.

Examples I to III and comparative example C1 and C2

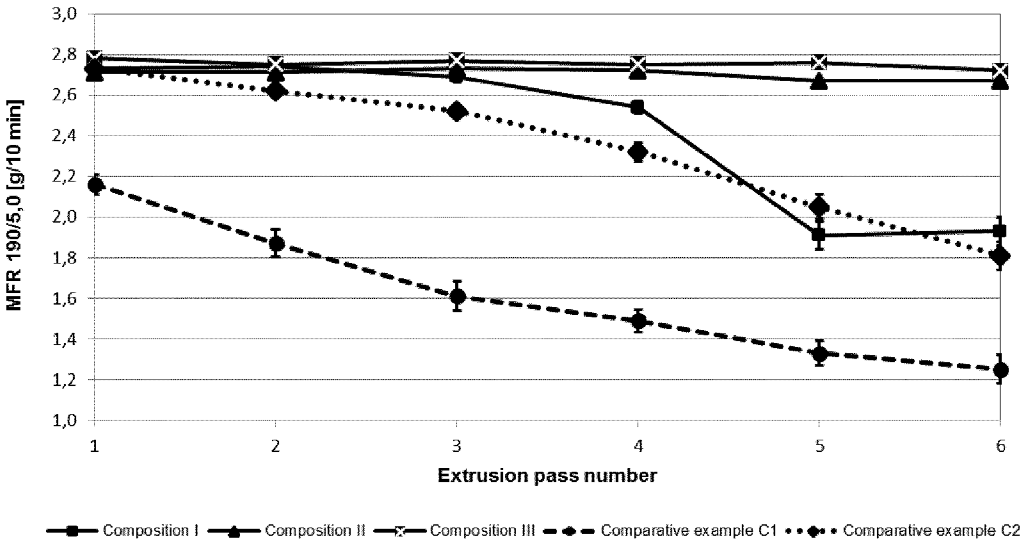
Table 1 shows a comparison between stabilized polyolefin compositions according to the invention (I, II and III) and comparative examples C1 and C2.

Table 1

		Concentration versus LLDPE
C1	No additives	0
I	α-tocopherol	50 ppm
Ib	α-tocopherol	75 ppm
II	α-tocopherol	100 ppm
IIb	α-tocopherol	150 ppm
III	α-tocopherol	200 ppm
C2	Irganox 1076 + Irgafos 168	200 ppm + 800 ppm

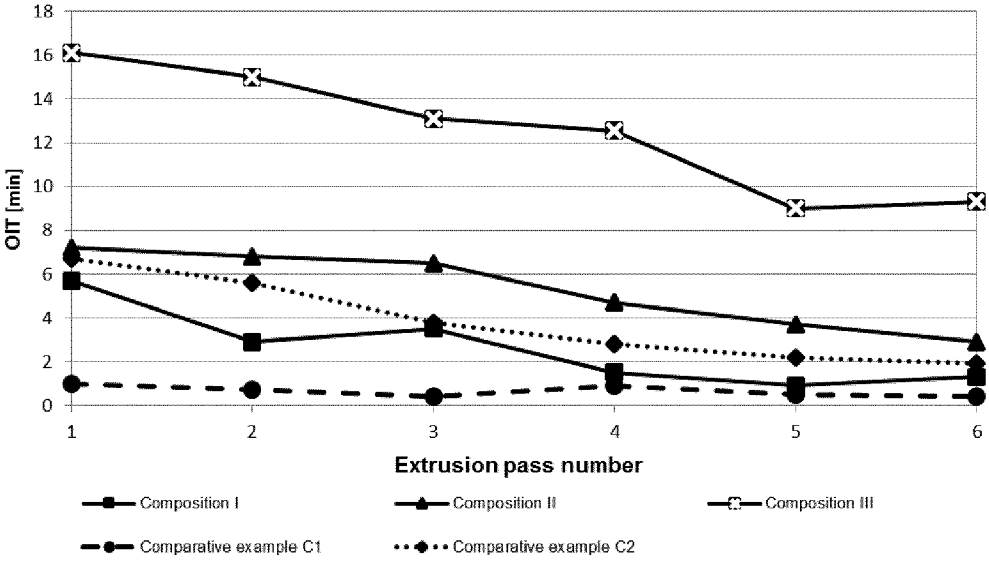
Irganox 1076 is thereby octadecyl 3,5-di-t-butyl-4-hydroxyhydrocinnamate and Irgafos 168 is thereby tris (2,4-di-t-butylphenyl) phosphite.

Fig.1



Comparison of compositions I to III with comparative examples C1 and C2 shows that processing stability for the compositions according to the invention is better than for C1 and/or comparable or better than for C2, in that the MFR does not significantly decrease after two consecutive extrusions, preferably even after 3 consecutive extrusions and for composition II and III even after 6 consecutive extrusions. This means that cross-linking occurring during extrusion may be reduced.

Fig. 2



Moreover, OIT can be increased for the compositions I to III compared to C1 and/or for compositions II and III even compared to C2. Furthermore, an increased OIT may also be maintained over several consecutive extrusions.

Table2

		Concentration versus LLDPE	Yellowness index after 3 rd extrusion	Yellowness index after 6 th extrusion
Ib	α-tocopherol	75 ppm	4.2	4.6

As shown above, the present invention may also lead to a low yellowness increase even after multiple extrusions.

CLAIMS

1. Stabilized polyolefin composition comprising
   - A. a copolymer of ethylene and at least one α-olefin, whereby the copolymer of ethylene and at least one α-olefin is linear low-density polyethylene (LLDPE) produced using at least one Ziegler-Natta catalyst,
   - B. between 50 and 200 ppm by weight of α-tocopherol relative to the component A.

2. Stabilized polyolefin composition according to Claims 1, wherein the composition further comprises:
   - C. a stearate

3. Stabilized polyolefin composition according to any one of Claims 1-2, wherein the linear low-density polyethylene (LLDPE) is produced using at least one Ziegler-Natta catalyst comprising Mg and at least one or one of Ti, Hf or Zr.

4. Stabilized polyolefin composition according to any one of Claims 1-3, wherein the amount of component B ranges between > 75 ppm and < 175 ppm by weight, further preferred between > 90 ppm and 170 or <170 ppm, further preferred between > 110 ppm and < 170 ppm and/or between > 50 ppm and <100 ppm, further preferred > 60 ppm and <90 ppm by weight, relative to the component A.

5. Stabilized polyolefin composition according to any one of Claims 2-4, wherein the amount of component C ranges between 100 ppm and 1000 ppm by weight relative to the component A.

6. Stabilized polyolefin composition according to any one of Claims 1-5, wherein the Mw/Mn of the linear low density polyethylene is between 4 and 12, preferably between 4.5 and 11, further preferred between 4.7 and 10, further preferred between 5 and 8, further preferred between > 5 and < 7, further preferred between > 5.5 and < 6.5 and/or the Mz/Mw of the linear low density polyethylene between 4 and 12, preferably between 4.5 and 11, further preferred between 4.7 and 10, further preferred between 5 and 8, further preferred between > 5 and < 7, further preferred between > 5.8 and < 6.8.

7. Stabilized polyolefin composition according to any one of Claims 1-6, wherein the low density polyethylene has a density of ≥ 850 kg/m^3 and ≤ 950 kg/m^3, preferably ≥ 910 kg/m^3 and ≤ 940 kg/m^3, further preferred between 913 kg/m^3 and 923 kg/m^3, further preferred between > 915 kg/m^3 and < 921 or < 920 kg/m^3 and/or a melt mass flow rate (MFR) as measured according to ISO 1131-1 (2011) at 190°C and at a load of 2.16 kg of between 0.1 dg/min and 10 dg/min, preferably between 0.5 dg/min and 5 dg/min, further preferred between 0.5 dg/min and 3 dg/min, even further preferred >0.5 dg/min and < 2 dg/min, even further preferred > 0.7 dg/min and < 1.5 dg/min.

8. Stabilized polyolefin composition according to any one of Claims 1-7, wherein the stabilized polyolefin composition does not comprise other tocopherols and/or comprises no tocotrienol and/or no natural vitamin E and/or no other antioxidant and/or no other compound comprising at least one phenolic motif and/or wherein the stabilized polyolefin composition comprises only α-tocopherol as antioxidant.

9. Stabilized polyolefin composition according to any one of Claims 1-8, wherein the increase of MFR between the 1st and the 6th extrusion is between ≥ 0 and <0.5 g/10min, preferably between ≥ 0 and <0.2 g/10min and/or the increase of yellowness index is between the 3rd and the 6th extrusion may be ≥ 0 and <0.7, preferably ≥ 0 and < 0.5, and/or the OIT after the 6th extrusion is between 2. 5 and 18 min.

10. An article prepared using the stabilized polyolefin composition according to any one of Claims 1-9.

11. A film prepared using the stabilized polyolefin composition according to any one of Claims 1-9.

12. A pouch prepared using the stabilized polyolefin composition according to any one of Claims 1-9.

13. A packaging prepared using the stabilized polyolefin composition according to any one of Claims 1-9.

14. Use of a stabilized polyolefin composition according to any one of Claims 1-9

for films and/or pouches for food and/or beverage packaging applications.

15. Use of a stabilized polyolefin composition according to any one of Claims 1-9 for biomedical and/or healthcare applications and/or for films and/or pouches for biomedical and/or health care, and/or pharmaceutical and/or medical applications.

INTERNATIONAL SEARCH REPORT

International application No
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A. CLASSIFICATION OF SUBJECT MATTER
INV. C08K5/1545
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)
C08K

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)

EPO-Internal, CHEM ABS Data, WPI Data

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	EP 0 878 502 A2 (CIBA GEIGY AG [CH]) 18 November 1998 (1998-11-18) page 3, lines 20,37; claims 1, 3, 5, 8, 9, 14; tables 2,5 page 14, line 49 -----	1-15

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☒ 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

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"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

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"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 1 March 2017	Date of mailing of the international search report 14/03/2017
Name and mailing address of the ISA/ European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Fax: (+31-70) 340-3016	Authorized officer Lohner, Pierre

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International application No
PCT/EP2016/082545

C(Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	17: Macromol. Symp, 1 January 2001 (2001-01-01), pages 17-29, XP055138079, Retrieved from the Internet: URL:http://onlinelibrary.wiley.com/store/10.1002/1521-3900(200112)176:1<17::AID-MASY17>3.0.CO;2-D/asset/17_ftp.pdf?v=1&t=hzmp8pxo&s=51ee5feeac837acbc29c0ea445af10e33e544234 [retrieved on 2014-09-03] paragraph [Summary]; figures 1-4 paragraph [Conclusions] -----	1,3,4, 6-15
X	EP 0 682 073 A2 (PHILLIPS PETROLEUM CO [US]) 15 November 1995 (1995-11-15) page 3, line 57; claims 1,2,6 page 2, line 7 -----	1,3,4, 6-10,13
X	WO 2014/190298 A1 (DOW GLOBAL TECHNOLOGIES LLC [US]) 27 November 2014 (2014-11-27) claim 1; examples IEC-1 to IEC-7; tables -----	1-10

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INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No
PCT/EP2016/082545

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