Title: PROCESS FOR THE MANUFACTURE OF ARTIFICIAL GRASS CARPETS

Abstract: The invention relates to a method for the manufacture of artificial grass which consists of spraying onto the rear of the primary turf carpet a two- or three-component polyurethane product, that crosslinks rapidly, even at low temperatures, to give a compact, elastic, uniform and bubble-free film of a coating-adhesive, which shows an improved chemical and water resistance and bonds firmly the tufts to the primary web. The invention discloses specific products for this method.
FIELD OF INVENTION

The present invention relates to a manufacturing process for artificial grass carpets.

PRIOR ART

Artificial grass is increasingly in demand for sporting applications such as soccer and five a side pitches, tennis courts, recreational applications such as play areas, and decorative applications in the residential and non-residential construction industry.

The drawback of natural compared with artificial grass carpets is its high cost of maintenance, due, for example, to the need for daily watering, weekly cutting in order to regularize its height, and the need to limit its use to areas with sufficient natural lighting.

For this reason, demand for artificial grass carpets turf is continually increasing.

In particular, some sports federations are beginning to allow its use instead of natural grass for both recreational and competitive events.

Artificial grass carpets are manufactured today using a filled SBR resin in aqueous dispersion, which is partially post-crosslinked at high temperature, or using two- or three-component cellular polyurethane (see for example US 5,104,693), which are crosslinked at high temperature too.

For example, in most cases, the back of the artificial grass consists of:

1) a perforated polypropylene primary carpet (different primary carpets are distinguished by the distance between the holes);

2) a non-woven needle-punched polypropylene or polyester/propylene fabric (fleece);
3) tufts made of PE/PP, PP or PE yarns (polyamide yarns will be used in future). These yarns are pre-oiled.

In most cases, the rear of artificial grass carpets is currently coated with an SBR latex based product 74% in solids, filled with CaCO₃, with a viscosity of 2-3000 mPas at 23°C and a consumption of 1.1 kg/sq.m equivalent to approx. 800 g/sq.m of dry matter.

The latex is applied to the carpet in one of two ways:

a. the roller on which the carpet runs is immersed in a latex bath;

b. the latex is distributed by a mobile applicator.

The excess material must then be removed with a doctor blade to regularize its thickness.

The drying of the latex takes place in a tunnel of ovens, usually 15-50 m long, in which the carpet remains for approx. 3-10 min, running at the rate of approx. 2-4 m/min. The heat cycle involves temperatures of between 120 and 150°C.

The function of the water-based product is to impregnate the substrate, thus fixing the tufts without compromising the dimensional stability of the carpet.

In other cases, the SBR-based aqueous product is replaced by a two- or three-component polyurethane product, which is similarly applied, the components being pre-mixed through a foaming machine. These last type of products guarantees superior characteristics of resilience.

For this reason, some new regulations require performances which can only be achieved by the use of said reactive polyurethane products.

The two or three components polyurethane product is distributed onto the rear of the turf carpet, coupled with a non-woven fabric, using a foaming machine and it is regularized by a doctor blade as used for the SBR latex.

The excess material must then be removed with a doctor blade to
regularize its thickness.

Both the residence times and the temperatures in the oven tunnel required by these polyurethane products are similar to those of products based on an SBR dispersion.

Furthermore if the manufacturing cycle has to be interrupted due to a technical problem, lengthy operational times for the cleaning of the line are required to avoid accumulations of hardened material.

For the same reason, if the manufacturer has to alternate the production of SBR-based products and polyurethane products, lengthy cleaning operations on several parts of the plant are required.

**DESCRIPTION OF THE INVENTION**

The object of the invention is a novel technology and novel specific polyurethane products for the manufacture of artificial grass.

The technology involves spraying onto the rear of the artificial grass carpet with a two- or three-component polyurethane product that crosslinks rapidly, even at low temperatures, to give a compact, elastic, uniform film of coating-adhesive, with no bubbles, which is resistant to water and bonds the tufts firmly to the synthetic primary turf carpet.

The two or three-component polyurethane product preferably consists of a system wherein part A comprises one or more polyols, fillers, catalysts and additives, etc., and part B consists of the isocyanate component.

Component A of the two-component polyurethane product preferably consists of one or more polyols, in particular diols and/or triols or polyether polyols obtained by polymerisation preferably of propylene oxide, with a molecular mass ≤ 5,000 Daltons in the proportion of 15% to 60%, and mineral fillers in the proportion of 85% to 40% by weight.

One of the polyols can be a vegetable oil or one of its derivatives; preferably it is the castor oil, in the percentage by weight of between 0% and
100% of the polyol mixture.

The mineral fillers of component A consist of carbonates, sulphates, silicates, oxides of alkaline earth metals, and other usual fillers, possibly combined with one another, calcium carbonate preferably being used as the main filler. Microspheres or any kind of scraps can also be used as fillers.

The additives, included in component A in minor proportions, may be organic or inorganic pigments, preferably carbon black, chemical or physical dryiers, antisetttling and antifoaming agents, antifungus, antioxidants or UV stabilizers, and catalysts.

Component A of the three-component polyurethane products has the same composition of that one described above for the two component polyurethane product apart the catalyst(s) which is introduced into the component C. The catalyst in component A of the two component polyurethane product is selected from tertiary amines and/or organometallic compounds in the proportion of 0.001 to 3% by weight of the polyurethane product.

The catalyst is preferably selected from triethylenediamine, morpholine, DBU (1,8-Diazabicyclo(5.4.0.)undec-7-ene), compounds of tin, bismuth, titanium and zirconium, preferably triethylenediamine or tin compounds such as tin dibutyldilaurate or di-n-butyltinbis(mercaptoaceticacidisooctyl ester).

In case of using organometallic catalysts it can be convenient to feed the catalyst (or a catalyst composition) as third component (part C) due to the limited storage stability of these catalysts in the part A.

In fact organometallic catalysts can decompose during the storage and affect important changes of the reactivity of the 2 component product.

The catalyst composition can be carried out if necessary by diluting the catalyst in a small amount of polyol.

The ratio of the components (A+B) to the component C can vary from
100 to 0.1 to 100 to 5 by weight.

Component B of the two-component polyurethane product typically consists of an isocyanate prepolymer obtained from one or more polyols, 4,4′ diphenylmethane diisocyanate, possibly mixed with isomers and homologues, or modified with 15-25% of carbodiimide, characterised by an NCO content of between 5 and 25%, preferably between 10 and 20%.

Alternately it consists of a MDI based polyisocianate with an NCO (%) content of between 25% and 33.5%.

The viscosity of the two-component polyurethane when parts A and B (without catalyst) are mixed is under 10,000 mPa.s at +50°C, and preferably ≤ 6000 mPa.s at +50°C.

The two component polyurethane product is sprayed with nozzles fed by pressurised pumps, and preferably by low-pressure gear pumps, which produce an A/B mixing ratio by weight of between 1:1 and 10:1.

Components A and B and - in the case of a three component polyurethane product - component C are fed separately and mixed at the spraying head through a static or dynamic mixer. The material is then sprayed evenly onto the rear of the primary artificial grass. Ratio and distribution are driven by a computer assisted equipment. Components A and B, or A, B and C penetrate deeply into the tufts of artificial grass, and react by bonding the yarns firmly to the primary turf carpet.

The use static mixers in cartridges minimises the quantity of mixed material which is liable to harden in the event of stoppage of the plant.

The specific products according to the invention can therefore be formulated at high reactivity, what is not possible in the conventional manufacturing processes.

The advantages of combined use of the technology and the specific two or three - polyurethane products according to the invention are:
1) the possibility of reducing the times and temperatures involved in the post-crosslinking process in the oven, and even its total elimination;

2) a considerable increase in the running speed of the carpet, and consequently of the industrial productivity per line;

3) compatibility (i.e. the possibility of coexistence) between the spraying technology and the spreading and calendering technology, if necessary;

4) the possibility of interrupting the spray application of the polyurethane coating-adhesive at any time, without any problems, what is not possible for a polyurethane coating-adhesive spread through a doctor blade;

5) elimination of the need to bond the non-woven fabric (fleece) to the primary web, which is required when aqueous adhesives are used, such as those based on SBR dispersions;

6) the possibility to reduce the concentration of volatile isocyanate monomer aerosols in the workplace if the component B is a MDI based isocyanate prepolymer with an NCO percentage of between 5 and 25%;

7) furthermore the oven curing temperature of 80-100°C allows to reduce the shrinking of the pile yarns; this result can't be achieved by the use of SBR technology which requires higher oven temperature (120-150°C).

Piston pumps and gear pumps, airless nozzles and nozzles with assisted air, are suitable to feed the two components A and B or the three component A, B and C.

Low pressure gear pumps with assisted air are preferable for continuous robotised processing.
Suitable gear pumps, mixing heads and computer assisted equipment for two or three-component polyurethane products are available by Maxver s.r.l. (San Cesario sul Panaro, Italy).

The running speed of the carpet on the production line should be at least 1 m/minute, preferably is between 2 and 15 m/min, and most preferable is between 6 and 12 m/min for consumption of between 500 and 1500 g/m², and preferably between 700 and 900 g/m².

Crosslinking can be performed at room temperature, but is normally carried out in a heated tunnel at temperatures of between +40 and +120°C, preferably between 80°C and 120°C, and more preferably between 80 and 100°C.

Under these conditions, high productivity is guaranteed with short oven residence times, leading to benefits in terms of energy consumption.

For the application of the spray technology the polyurethane product (without catalyst) must have a viscosity of less than approx. 10,000 mPa.s, and preferably less than 6000 mPa.s, measured at the temperature of use.

The application temperature should be between +15 and +80°C, and preferably between +30 and +65°C.

If said conditions are maintained, excellent penetration of the adhesive into the perforated primary turf carpet (based on polypropylene, for example) will be achieved, and the artificial grass (based on, for example, polypropylene, polyethylene, polyethylene/polypropylene, polyamide, etc.) will present an very good tuft lock.

The use of the novel application technology and the novel polyurethane carpetbacking gives to the end products very good properties of water and chemical resistance and fungal growth resistance too.

EXAMPLE 1

A two-component polyurethane product consists of:
Part A: Filled polyol
1) Castor oil FSG (AlberdingK-Boley) pp 20.0
2) Desmophen 1380 BT (Bayer) pp 4.3
3) Calcit GS 15 (Ros) pp 71.67
4) Dabco T12 (Air Products) pp 0.03
5) Sylosiv A4 (Grace) pp 4.0

Ingredients 3, 4 and 5 are gradually added to ingredients 1 and 2 in a disperser, until dispersion is complete.

Viscosity: 120,000 mPa.s at 23°C

Specific gravity 1.8 g/cc

Part B: Isocyanate prepolymer
1) Desmodur LS 2424 (Bayer) pp 11.6
2) Desmodur 44 MC (Bayer) pp 23.15
3) Desmodur CD-S (Bayer) pp 3.86
4) Desmophen 2061 BD (Bayer) pp 38.6
5) Suprasec 2029 (Huntsman) pp 22.81

Ingredients 1 and 3 are fed, in that order, into a steel reactor equipped with a stirrer, operating in a dry nitrogen atmosphere. Under stirring, the temperature of the solution is raised from room temperature to 40-45°C.

Ingredients 2 and 4 are then fed in, in that order, the temperature is raised to +60°C, and stirring is continued for 6 hours.

After this stage, the temperature is reduced to +50°C and ingredient 5 is fed in, stirring for 15 min, until homogenisation is complete.

% NCO = 16.8%

Viscosity = approx. 1200 mPas at +23°C

Weight ratio A/B = 4/1

Viscosity A+B= approx. 5000 mPas at +50°C (measured without catalyst)
Part A and Part B are pre-heated at the temperatures of +80°C and +35°C respectively and transferred, via positive-displacement gear pumps (Dosamix 2C.S. made by Maxver s.r.l., San Cesario sul Panaro, Italy) and thermoheated tubes, to the spray gun, which operates with low-pressure compressed air and it is equipped with a static mixer.

The two-component polyurethane product is sprayed regularly and evenly onto the rear of a polypropylene primary turf carpet (type Soccerpro Diamond from Limontasport, Cologno al Serio, Bergamo, Italy) without fleece, which runs at 4 m/min at the consumption rate of approx. 850 g/sq.m.

Crosslinking is completed in the oven after six min at 80°C.

At the exit of the oven the product is perfectly crosslinked, and the turf presents a tuft lock, after 2 days at +23°C, of about 5 daN (according to ISO 4919).

The coverage of the artificial grass loops and the penetration between the yarns are optimal.

After 6 min at 80°C and 2 days at 23°C, a 1 mm thick film of product (applied simultaneously in line in a mould) presents:

- elongation at break: 150% (according to DIN 53504),
- tear strength: 14.5 N/mm (according to ISO 34-1),
- Shore Hardness A: 60 (according to DIN 53505).

The environmental impact of the product was evaluated, according to OSHA method no. 47, by monitoring the MDI monomer in the atmosphere, the sampling points being located at a distance of 0.5 and 1.0 m from the spray gun in the spraying area (zone A) and 4 m far from the spray gun immediately before the entry into the oven (zone B), with the following results:

Zone A - MDI monomer: 24.7 µg/cu.m
Zone B - MDI monomer: 16.9 µg/cu.m

The TLV of MDI is 51 µg/cu.m.
A test for diaminodiphenylmethane was also conducted in the same area and on the chimney of the plant. Two Carbotrap tubes in series were used for this purpose; the products of thermal desorption were then analysed by gas chromatography.

The aromatic amine was not detected in any case.

The storage stability of the component A (containing Dabco T12) is limited to one month at room temperature. In order to get longer storage stability this composition should be sprayed using the three component equipment as above mentioned.

**EXAMPLE 2**

A two-component polyurethane product consists of:

**Part A:** Filled polyol

1) Castor oil FSG (AlberdingK-Boley) pp 20.0
2) Desmophen 1380 BT (Bayer) pp 2.8
3) Omyacarb 15 AV (Omya) pp 71.6
4) Nero Spezial 4 (Degussa) pp 0.1
5) Dabco 33 LV (Air Products) pp 1.5
6) Sylosiv A4 (Grace) pp 4.0

Ingredients 3, 4 and 5 are gradually added to ingredients 1 and 2 in a disperser, until dispersion is complete.

Viscosity: 60,000 mPas at 23°C

Specific gravity 1.8 g/cc

**Part B:** Isocyanate prepolymer

1) Isonate 143 M (Dow) pp 57.2
2) Desmophen 2061 BD (Bayer) pp 42.8

Ingredients 1 and 2 are fed into a steel reactor equipped with a stirrer, operating in a dry nitrogen atmosphere. Under stirring, the temperature of the solution is raised from room temperature to +60°C, and stirring is continued
for two hours.

After this stage, the temperature is reduced to +50°C

% NCO = 15%

Viscosity = approx. 1700 mPas at +23°C

Weight ratio A/B = 4/1

Viscosity A+B= approx. 4000 mPas at +50°C (measured without catalyst).

The reactivity of this two component polyurethane composition containing Dabco 33LV doesn't change after more than one month storage at 50°C.

The two component polyurethane product, sprayed on a mold using the same laboratory equipment indicated in the example 1 at 1 mm thickness, shows after a curing of three min at 100°C following properties:

- tensile strength: 2.8 N/mm²
- elongation at break: 170% (according to DIN 53504)
- tear resistance: 12.7 N/mm (according to ISO34-1)
- shore Hardness A: 50 (according to ISO 53505)
- flexibility at 0°C, on a cylindrical mandrel (5 mm diameter): OK

The two component polyurethane product, applied at 850 g/sq.m on the rear of a polypropylene primary turf carpet (we use the same Soccerpro Diamond from Limontasport indicated in example 1) shows after curing of three min at 100°C following properties:

- tuft lock: 5.8 daN (according to ISO 4919)
- water resistance after 50 days at 23°C of the artificial grass (expressed as percentage variation by weight):
- resistance to fungal growth (test method SM 022 a) carried out on free film of polymer:

<table>
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<th></th>
<th>deionised water</th>
<th>acid water (H₂O saturated with CO₂)</th>
<th>alkaline water (20% KOH in H₂O)</th>
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<td>weight variation</td>
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<td>Polyurethane Mapei</td>
<td>+7%</td>
<td>+7%</td>
<td>+12%</td>
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<tr>
<td>SBR</td>
<td>+19%</td>
<td>+19%</td>
<td>+21%</td>
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<tr>
<td>Cellular PU competitor</td>
<td>+28%</td>
<td>+28%</td>
<td>+25%</td>
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Legend:

0 = from plate completely free of growth up to growth only on the edge of the sample (max 1%)

1 = growth from the edge (up to 25%)

5
1. A process for manufacturing artificial grass carpets wherein the tufts of the artificial grass are fixed to the primary turf carpet by spraying a two or three-component polyurethane product able to crosslink rapidly, even at low temperatures, to give a compact, elastic, uniform film of an adhesive-coating, which is resistant to water.

2. Manufacturing process as claimed in claim 1, wherein the spray application is carried out on the rear of the primary turf carpet without the use of non-woven fabric (fleece).

3. Manufacturing process as claimed in claim 1 or 2, wherein the two-component polyurethane product is solvent-free and comprises a first component A consisting of at least one polyol, fillers, catalysts and other conventional components, and a second component B consisting of an isocyanate prepolymer, or of a MDI based polyisocyanate.

4. Manufacturing process according to claims 1 and 2 wherein the three-component solvent-free polyurethane product comprises a first component (A) consisting of at least one polyol, fillers and other conventional components, a second component (B) consisting of an isocyanate prepolymer or a MDI based polyisocyanate, and a third component (C) consisting of a catalyst composition.

5. Manufacturing process as claimed in any one of claims 1 to 4, wherein component A of the two- or three-component polyurethane product consists of one or more polyols with a molecular mass of ≤ 5,000 Daltons in the proportion of 15% to 60%, and mineral fillers in the proportion of 85% to 40% by weight.

6. Process as claimed in claim 5, wherein the polyols are diols and/or triols or polyether polyols preferably obtained by polymerisation of propylene
oxide.

7. Manufacturing process as claimed in claim 3, 4, 5 or 6 wherein one of the polyols can be a vegetable oil, or one of its derivatives thereof, preferably is castor oil, in the percentage by weight of between 0% and 100% of the polyol mixture.

8. Manufacturing process as claimed in any one of claims 3 to 7, wherein the fillers of component A consist of carbonates, sulphates, silicates, oxides of alkaline earth metals, pigments like carbon black, microspheres and any kind of scraps, calcium carbonate preferably being used as the main filler.

9. Manufacturing process as claimed in any one of claims 3 to 8, wherein the catalyst(s) contained in component A, or separately added as component C, is (are) selected from tertiary amines and/or organometallic compounds and is (are) fed in the proportion of 0.001 to 3% by weight of the polyurethane product.

10. Manufacturing process as claimed in claim 9, wherein the catalyst is selected from triethylenediamine, morpholine, DBU, compounds of tin, bismuth, titanium and zirconium, preferably triethylenediamine or tin compounds such as tin dibutyldilaurate or di n-butylbis(mercaptoacetic acid isoocyt ester).

11. Manufacturing process as claimed in any one of claims 1 and 3 to 10, wherein the two- or three-component polyurethane product comprises a component B consisting of an isocyanate prepolymer obtained from one or more polyols, 4,4' diphenylmethane diisocyanate, possibly mixed with isomers and homologues, and/or modified with 15-25% carbodiimide and/or modified with allophanate groups, characterised by an NCO content of between 5 and 25%, preferably between 10 and 20%.

12. Manufacturing process as claimed in any one of claims 1 and 3 to 10, wherein in the two- or three-component polyurethane product the component
B consists on a MDI based polyisocyanate with an NCO (%) content of between 25% and 33.5%.

13. Manufacturing process as claimed in any one of claims 3 to 12, wherein the viscosity (measured without catalyst) of the polyurethane product is under 10,000 mPa.s. at +50°C, and preferably ≤ 6000 mPa.s at +50°C.

14. Manufacturing process as claimed in any one of claims 3 to 13, wherein the tufts of grass are fixed directly to the back of the carpet with 500-1500 g/m², and preferably 700-900 g/m², of the two-component polyurethane product.

15. Manufacturing process as claimed in any one of claims 3 to 14, wherein the polyurethane product is sprayed at temperatures of between 15 and 80°C, preferably between 30 and 65°C, with nozzles fed by pressurised pumps and preferably by low-pressure air assisted gears pumps which produce an A/B mixing ratio by weight of between 1:1 and 10:1 or an (A+B)/C mixing ratio by weight of between 100:0.1 to 100:5.

16. Manufacturing process as claimed in any one of claims 3 to 15, wherein the polyurethane product applied to the rear of the primary turf carpet is subjected to post-crosslinking in the oven at temperatures of between 40 and 120°C, and preferably between 80 and 100°C.

17. Manufacturing process as claimed in any one of claims 3 to 16, which can be performed continuously with oven residence times of at least 1 minute.

18. Solvent-free two-component polyurethane product for use in the process disclosed in claims 1-16, comprising a first component A consisting of at least one polyol, fillers, other conventional components and catalysts, and a second component B consisting of an isocyanate prepolymer or of an MDI based polyisocyanate.

19. Solvent free three-component polyurethane product for use in claims 1-18 comprising a first component A consisting of at least one polyol, fillers
and other conventional components, a second component B consisting of an isocyanate prepolymer or of a MDI based polyisocianate, and a third-component C consisting of a catalyst(s) composition.

20. Product as claimed in claim 18 and 19, wherein component A consists of one or more polyols with a molecular mass of ≤ 5,000 Daltons in the proportion of 15% to 60%, and mineral fillers in the proportion of 85% to 40% by weight.

21. Product as claimed in claim 17, 18, 19 or 20, wherein the polyols are diols and/or triols or polyether polyols obtained preferably by polymerisation of propylene oxide.

22. Product as claimed in claim 18, 19, 20 or 21, wherein one of the polyols is a vegetable oil or one of its derivatives, preferably it is castor oil, in the percentage by weight of between 0% and 100% of the polyol mixture.

23. Product as claimed in any one of claims 18 to 22, wherein the fillers of component A consist of carbonates, sulphates, silicates, oxides of alkaline earth metals, pigments like carbon black, microspheres and any kind of scraps, calcium carbonate being preferably used as the main filler.

24. Product as claimed in any one of claims 18 to 23, wherein the catalyst contained in component A of the two-component polyurethane product or in component C of the three-component product is selected from tertiary amines and/or organometallic compounds and calculated in the proportion of 0.001 to 3% by weight of the polyurethane product.

25. Product as claimed in claim 24, wherein the catalyst is selected from triethylenediamine, morpholine, DBU, compounds of tin, bismuth, titanium and zirconium, preferably triethylenediamine or tin compounds such as tin dibutyldilaureate or di n-butylbis(mercaptoaceticacid isooyctyl ester).

26. Product as claimed in any one of claims 18 to 25, comprising a component B consisting of an isocyanate prepolymer obtained from one or
more polyols, 4,4' diphenylmethane diisocyanate, possibly mixed with isomers and homologues, and/or modified with 15-25% carbodiimide, and/or modified with allophanate groups, characterised by an NCO content of between 5 and 25%, most preferably between 10 and 20%.

27. Product according to claims 15 to 25, wherein the component B of two- or three-component polyurethane product consist of a MDI based polyisocyanate with NCO (%) content of between 25% and 33.5%.

28. Product as claimed in any one of claims 18 to 27, wherein the viscosity of the two or three-component polyurethane product without catalyst is under 10,000 mPas at +50°C, and preferably ≤ 6000 mPas at +50°C.
A. CLASSIFICATION OF SUBJECT MATTER

INV. D06N3/14 D06N7/00 E01C13/08

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)
D06N E01C

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

Electronic database consulted during the international search (name of database and, where practical, search terms used)
EPO-Internal, WPI Data

C. DOCUMENTS CONSIDERED TO BE RELEVANT

<table>
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<th>Citation of document, with indication, where appropriate, of the relevant passages</th>
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<tr>
<td>X</td>
<td>US 3 730 822 A (LOVELL J ET AL) 1 May 1973 (1973-05-01) col umn 2, lines 38-41; cl aims 1,6,8; examples column 3, line 64 - column 4, line 4 column 6, lines 20-31 column 6, line 59 - column 7, line 20 column 8, lines 38-47</td>
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X Further documents are listed in the continuation of Box C.

X See patent family annex.

Special categories of cited documents:

1A: document defining the general state of the art which is not considered to be of particular relevance

1E: earlier document but published on or after the international filing date

1L: 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)

1O: document referring to an oral disclosure, use, exhibition or other means

1P: document published prior to the international filing date but later than the priority date claimed

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

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

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

*8: document member of the same patent family

Date of the actual completion of the international search
19 February 2009

Date of mailing of the international search report
03/03/2009

Name and mailing address of the ISA/
European Patent Office, P.B. 5818 Patentlaan 2
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Tel. (+31-70) 340-2040, Fax (+31-70) 340-3016

Authorized officer
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<td>US 2002/074073 AI (HAMRICK GLEN [US] ET AL) 20 June 2002 (2002-06-20) paragraphs [0002], [0022], [0023], [0024], [0029], [0030], [0031] and [0033] claims 1, 3-5</td>
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<td>X</td>
<td>WO 00/37737 A (BAYER AG [DE] ; LYONDELL CHEMIE TECHNOLOGIE NE [NL]) 29 June 2000 (2000-06-29) page 10, lines 20,21, 28-30; claim 1; examples 1-6, C1-C6; tables 1, 3, 4 page 15, line 12</td>
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<td>X</td>
<td>WO 98/08893 A (DOW CHEMICAL CO [US]; JENKINES RANDALL C [US]) 5 March 1998 (1998-03-05) page 11, lines 6-28; example 1</td>
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<td>P,X</td>
<td>WO 2008/073808 A (DOW GLOBAL TECHNOLOGIES INC [US]; JENKINES RANDALL [GE]; PERRY THOMAS) 19 June 2008 (2008-06-19) page 4, lines 16-19, 24, 25; claims 6, 7; examples 4, 6 page 5, line 1 page 6, paragraph 4 page 8, lines 6-8, 21-24 page 9, paragraph 2 page 10, lines 15, 33-36 page 11, lines 1-3</td>
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<td>X</td>
<td>US 3 895 149 A (SHEFFLER ROBERT H ET AL) 15 July 1975 (1975-07-15) column 3, lines 16, 17, 27-33; claim column 5, lines 56-60 column 6, lines 10, 15-22, 53-66 column 7, lines 55, 56 column 8, lines 17-25, 45-58 column 9, lines 6-20</td>
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