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(54) Title: "PROCESS FOR THE DMC-CATALYZED PREPARATION OF POLYOLS"

(57) Abstract: The present invention relates to an improved process for preparing polyether alcohols by means of DMC catalysis, to the polyether alcohols prepared with the process, and to their use, in particular for producing polyurethanes.

### **Process for the DMC-catalyzed preparation of polyols**

The present invention relates to an improved process for preparing polyether alcohols by means of DMC catalysis, to the polyether alcohols prepared with the process, and to their use, in particular for producing polyurethanes.

The preparation of polyether alcohols by means of DMC catalysis is now prior art, especially for polyols for slabstock and CASE polyols. The advantages of the DMC catalysts are firstly that the addition reaction of the alkylene oxides proceeds at a higher reaction rate, and secondly the polyether alcohols prepared in this way are characterized by a lower content of unsaturated fractions in the polyether chain.

The DMC catalysts can be used particularly advantageously in the preparation of polyether alcohols with high molecular weight and low functionality, as are used in particular for producing polyurethane flexible foams. To produce polyurethane rigid foams, in most cases polyether alcohols with a high functionality, in particular those with a functionality of at least 4, are used.

In practice, however, in the production process of the polyols, fluctuations in the process parameters and the product quality are found time and again, especially an extension of the initiation time during the synthesis and an increased viscosity of the polyols. This may result in a decrease in activity of the catalysts and thus to a partial

or even complete prevention of the reaction of the alkylene oxides. The reduced activity of the catalysts can lead not only to uneconomical process control, but also to conditions in the alkoxylation reactor that are unacceptable from a safety point of view. Furthermore, the amount of catalyst and/or the reaction temperature must be increased in order to compensate for the drop in activity. Whereas small amounts of catalyst may remain in the product depending on the further use of the polyether alcohols, in the case of a reaction procedure with an increased amount of catalyst, costly purification steps have to be carried out to remove the catalyst constituents. Moreover, as a result of the reduced catalyst activity, polyether alcohols of lower quality, i.e. with a relatively high fraction of impurities and/or by-products, in particular low molecular weight compounds, may be formed and these can only be separated off in a very complex manner, if at all. The further processing of such polyether alcohols to polyurethanes and/or flexible or rigid foams consequently likewise leads to products of lower quality, which may be evident for example from gas evolution of the by-products present in the polyether alcohols, in particular low molecular weight compounds (for example formaldehyde).

The object of the invention was to improve the process such that these disturbances no longer occur.

It has now been found that the problem can be solved by using propylene oxide (PO) which comprises certain substances in concentrations which are lower than a certain threshold value for the alkoxylation in the preparation process of the polyols.

It has been found that a series of troublesome substances in PO considerably influences the initiation time and the properties of the resulting polyols. These are:

- a) chlorine compounds and bromine compounds;
- b) aldehydes (e.g. acetaldehyde) and ketones (e.g. acetone);
- c) allyl alcohol;
- d) hydrocarbons, for example lower saturated and unsaturated hydrocarbons;
- e) water and methanol;
- f) acids, for example carboxylic acids such as acetic acid.

The invention provides a process for preparing polyols by means of the addition reaction of alkylene oxides onto H-functional starter molecules in the presence of DMC catalysts, wherein, during the alkoxylation, propylene oxide is used in which the sum of the contents of organo-halogen compounds, in particular chlorine compounds and bromine compounds, determined according to DIN 51408-2 and EN ISO 10304, is not more than 150 ppm, for example not more than 60 ppm, or even not more than 30 ppm; in which the sum of the contents of aldehydes and ketones, determined according to VDI 3862 Part 2, is not more than 300 ppm, for example not more than 120 ppm, or even not more than 60 ppm; whose content of allyl alcohol, determined by means of gas chromatography analogously to DIN 51405, is not more than 8500 ppm, for example not more than 3400 ppm, or even not more than 1700 ppm; whose content of hydrocarbons, the content of which is determined by means of gas chromatography analogously to DIN 51405 and is expressed as the sum value, is not more than 500 ppm, for example not more than 200 ppm, or even not more than 100 ppm.

In one embodiment of the process, propylene oxide is used whose content of water, determined by Karl-Fischer titration according to DIN EN 13267, is not more than 500 ppm, for example not more than 200 ppm, or even not more than 100 ppm.

In a further embodiment of the process, propylene oxide is used whose content of acid, expressed by the value determined by means of acid-base titration according to DIN EN 62021-2 or DIN 12634 and calculated for acetic acid, is not more than 100 ppm, for example not more than 40 ppm, or even not more than 20 ppm.

In a further embodiment of the process, propylene oxide is used whose content of ethylene oxide, determined by gas chromatography analogously to DIN 51405, is not more than 100 ppm.

The invention also provides the polyols prepared with the process according to the invention and also the use thereof for producing polyurethanes, for example polyurethane foams or polyurethanes for coatings, adhesives, sealing compositions or elastomers (CASE).

Polyurethanes are produced in large amounts. They are usually produced by reacting polyisocyanates with compounds having hydrogen atoms that are reactive towards isocyanate groups, in particular polyether alcohols and/or polyester alcohols.

Polyester alcohols are in most cases prepared by reacting polyfunctional alcohols with polyfunctional carboxylic acids. Polyfunctional alcohols which may be used are the polyols prepared according to the invention.

The preparation of polyether alcohols takes place according to the invention by the addition reaction of alkylene oxides onto H-functional starter molecules in the presence of DMC catalysts. H-functional starter substances which may be used are, for example, alcohols, in particular alcohols having 1 to 8 hydroxyl groups, preferably aliphatic and cycloaliphatic alcohols having 2 to 8 carbon atoms in the branched or unbranched alkyl chain or in the cycloaliphatic backbone.

In one embodiment, the polyfunctional alcohols are selected from the group comprising glycerol, trimethylolpropane, pentaerythritol, di- and tripentaerythritol, ethylene glycol and its homologs, in particular ethylene glycol and/or diethylene glycol, propylene glycol and its higher homologs, in particular propylene glycol and/or dipropylene glycol, 1,3-propanediol, 1,2-, 1,3-, 2,3- and 1,4-butanediol, pentane- and hexanediols, in particular 1,5-pentane-diols and 1,6-hexanediol.

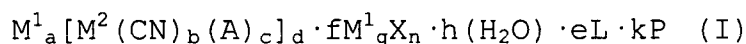
Further possible starters are hydroxycarboxylic acids, hydroxyaldehydes, hydroxyketones; tridecanol N and polymers thereof; esters of acrylic acid and methacrylic acid with difunctional alcohols such as HEA, HPA, HEMA, HPMA; vinyl ethers such as HBVE; isoprenol; polyesterols; lower alkoxylates of the aforementioned starters, of sucrose or sorbitol. The starters can be introduced as initial charge or, if appropriate, also be metered in during the process.

Alkylene oxides which can be used are, for example, ethylene oxide, propylene oxide, 1,2- or 2,3-butylene oxide, 1,2-isobutylene oxide, and also any desired mixtures thereof. In one embodiment of the invention, ethylene oxide, propylene oxide or mixtures thereof are used. In a further embodiment, exclusively propylene oxide is used.

The alkylene oxides can be added onto the starter molecules individually in the form of so-called blocks and, in the event of using different alkylene oxides, in any desired mixing ratio as mixed blocks. Moreover, the mixing ratio of the alkylene oxides during the synthesis can be varied discontinuously and also continuously.

Suitable DMC catalysts are described, for example, in EP 0 761 708 A2 and in EP 0 862 947 A1, both of which are hereby incorporated into this application by reference.

The double metal cyanide catalysts (DMC catalysts) satisfy the formula:



in which

$M^1$  is a metal ion selected from the group consisting of  $Zn^{2+}$ ,  $Fe^{2+}$ ,  $Co^{3+}$ ,  $Ni^{2+}$ ,  $Mn^{2+}$ ,  $Co^{2+}$ ,  $Sn^{2+}$ ,  $Pb^{2+}$ ,  $Mo^{4+}$ ,  $Mo^{6+}$ ,  $Al^{3+}$ ,  $V^{4+}$ ,  $V^{5+}$ ,  $Sr^{2+}$ ,  $W^{4+}$ ,  $W^{6+}$ ,  $Cr^{2+}$ ,  $Cr^{3+}$ ,  $Cd^{2+}$ ,  $La^{3+}$ ,  $Ce^{3+}$ ,  $Ce^{4+}$ ,  $Eu^{3+}$ ,  $Mg^{2+}$ ,  $Ti^{3+}$ ,  $Ti^{4+}$ ,  $Ag^+$ ,  $Rh^{1+}$ ,  $Ru^{2+}$ ,  $Ru^{3+}$ ,  $Pd^{2+}$ ;

$M^2$  is a metal ion selected from the group consisting of  $Fe^{2+}$ ,  $Fe^{3+}$ ,  $Co^{2+}$ ,  $Co^{3+}$ ,  $Mn^{2+}$ ,  $Mn^{3+}$ ,  $V^{4+}$ ,  $V^{5+}$ ,  $Cr^{2+}$ ,  $Cr^{3+}$ ,  $Rh^{3+}$ ,  $Ru^{2+}$ ,  $Ir^{3+}$ ;

and  $M^1$  and  $M^2$  are different;

A is an anion selected from the group consisting of halide, hydroxide, sulfate, carbonate, cyanide, thiocyanate, isocyanate, cyanate, carboxylate, oxalate and nitrate;

X is an anion selected from the group consisting of halide, hydroxide, sulfate, carbonate, cyanide, thiocyanate, isocyanate, cyanate, carboxylate, oxalate and nitrate;

L is a water-miscible ligand selected from the group consisting of alcohols, aldehydes, ketones, ethers, polyethers, esters, polyesters, polycarbonates, ureas, amides, nitriles and sulfides and mixtures thereof;

P is an organic additive selected from the group consisting of polyethers, polyesters, polycarbonates, polyalkylene glycol sorbitan esters, polyalkylene glycol glycidyl ethers, polyacrylamide, poly(acrylamide-co-acrylic acid), polyacrylic acid, poly(acrylamide-co-maleic acid), polyacrylonitrile, polyalkyl acrylates, polyalkyl methacrylates, polyvinyl methyl ethers, polyvinyl ethyl ether, polyvinyl acetate, polyvinyl alcohol, poly-N-vinylpyrrolidone, poly(N-vinylpyrrolidone-co-acrylic acid), polyvinyl methyl ketone, poly(4-vinylphenol), poly(acrylic acid-co-styrene), oxazoline polymers, polyalkyleneimines, maleic acid and maleic anhydride copolymers, hydroxyethylcellulose, polyacetates, ionic surface-active and interface-active compounds, bile acids and salts, esters and amides thereof, carboxylic acid esters of polyhydric alcohols and glucosides;

a, b, d, g and n are integers or fractions greater than zero, c, f, e, h and k are integers or fractions greater than or equal to zero, where at least one number is different from zero; with the proviso that a, b, c and d, and also g and n are selected such that the compound is electrically neutral.

In one embodiment, only f and k are zero if c is not zero and A is carboxylate, oxalate or nitrate.



The catalyst may be crystalline or amorphous. If  $k$  is zero, crystalline double metal cyanide compounds are preferred. If  $k$  is greater than zero, both crystalline, partially crystalline and also essentially amorphous catalysts are preferred.

In one embodiment, the catalyst of the formula (I) has a value for  $k$  which is greater than zero. The catalyst then comprises at least one multimetal cyanide compound, at least one organic ligand  $L$ ; and at least one organic additive  $P$ .

In another embodiment,  $k$  is zero or assumes a value such that the content of component  $P$  is not more than 4% by weight, based on the compound of the formula (I). If desired,  $e$  is likewise zero and  $X$  is exclusively carboxylate, preferably formate, acetate or propionate.

In another embodiment,  $f$  is greater than zero, preferably zero to 6. In this embodiment, in which one metal salt is present in the DMC catalyst, the catalyst has a structure which exhibits a particularly high catalytic activity. In this embodiment, the catalyst preferably has a crystalline structure.

In another embodiment,  $f$ ,  $e$  and  $k$  are different from zero. In this case, the DMC catalysts comprise a water-miscible organic ligand, mostly in an amount of from 0.5 to 30% by weight, and an organic additive, for example a polyether alcohol, mostly in an amount of from 5 to 80% by weight, as described e.g. in WO 98/16310 A1, which is hereby incorporated into this application by reference.

In one embodiment, the DMC catalyst is crystalline and has a platelet-like morphology. Such catalysts are described in WO 00/74845, which is hereby incorporated into this application by reference.

In one embodiment,  $M^1$  is  $Zn^{2+}$  and  $M^2$  is  $Co^{3+}$ , i.e. the multimetal cyanide compound is a zinc hexacyanocobaltate. Particular preference is given to crystalline multimetal cyanide compounds in which the anion X is formate, acetate or propionate and f is greater than 0, and which exhibit X-ray diffractograms as are described in DE 197 42 978 A1, which is hereby incorporated into this application by reference. Among these compounds, preference is in turn given to those in which the anion X is acetate and in particular those which crystallize in a monoclinic crystal system.

Crystalline DMC catalysts which can be used in the process according to the invention are also described in US 6,303,833 B1, US 6,613,714 B2, US 6,689,710 B2, US 7,470,823 B2, US 2004/249221 A1, US 2005/0203274 A1, and US 2008/0300376 A1, which are all incorporated into this application by reference.

The use of DMC catalysts has the advantage that the addition reaction of alkylene oxides proceeds at a relatively high reaction rate and the formation of undesired by-products is reduced compared to alkali metal hydroxide catalysis.

Processes for polyether alcohol synthesis by means of DMC catalysis are known and are described, for example, in EP 0 862 947 B1, DD 203 735 A1 or WO 99/51661 A1, which are all incorporated into this application by reference. The process can be carried out in a semibatch process, as

described e.g. in EP 1 259 560 A1, or else continuously, as in EP 0 912 625 A1, both of which are hereby incorporated into this application by reference.

The heat of reaction which forms during the alkoxylation can be dissipated either by an internal heat exchanger or else by an external one, or by means of a combination of the two. In the case of an external heat exchanger, the reaction contents are fed to and discharged from the heat exchanger by pumped circulation.

In the propylene oxide used in the alkoxylation, the sum of the contents of organohalogen compounds, in particular chlorine compounds and bromine compounds, determined according to DIN 51408-2 and EN ISO 10304, is not more than 150 ppm, for example not more than 60 ppm, or even not more than 30 ppm; the sum of the contents of aldehydes and ketones, determined according to VDI 3862 Part 2, is not more than 300 ppm, for example not more than 120 ppm, or even not more than 60 ppm; the content of allyl alcohol, determined by means of gas chromatography analogously to DIN 51405, is not more than 8500 ppm, for example not more than 3400 ppm, or even not more than 1700 ppm; and the content of hydrocarbons, determined by means of gas chromatography analogously to DIN 51405 and expressed as the sum value, is not more than 500 ppm, for example not more than 200 ppm, or even not more than 100 ppm.

In one embodiment, the propylene oxide has a halogen content of not more than 60 ppm, a content of aldehydes and ketones of not more than 120 ppm, a content of allyl alcohol of not more than 3400 ppm and a hydrocarbon content of not more than 200 ppm.

In a further embodiment, the propylene oxide has a halogen content of not more than 30 ppm, a content of aldehydes and ketones of not more than 60 ppm, a content of allyl alcohol of not more than 1700 ppm and a hydrocarbon content of not more than 100 ppm.

In one embodiment of the process, the propylene oxide has a water content, determined by Karl-Fischer titration according to DIN EN 13267, of not more than 500 ppm, for example not more than 200 ppm, or even not more than 100 ppm.

In a further embodiment of the process, the propylene oxide has an acid content, expressed by the value determined by means of acid-base titration according to DIN EN 62021-2 or DIN 12634 and calculated for acetic acid, of not more than 100 ppm, for example not more than 40 ppm, or even not more than 20 ppm.

Propylene oxide suitable for use in the process according to the invention can be obtained, for example, by subjecting propylene oxide prepared by means of the epichlorohydrin process, the SM/PO process, the HPPO process or the MTBE/PO process to a purification process in which the content of the aforementioned compounds in the propylene oxide is reduced to concentrations below the critical threshold values.

For example, impurities can be removed by extractive distillation of the propylene oxide, as is described, for example, in US 2005/082159 A, US 2003/102206, US 5,958,192 A, US 5,133,839 A, EP 0 685 471 A1 or EP 0 540 225 A1, which are all hereby incorporated into the present application by reference.

Another option is the treatment of the propylene oxide with zeolites, as arises for example from JP 2003/160573 A, which is hereby incorporated into the present application by reference.

A further option is the extraction with suitable extractants, as arises for example from EP 0 512 777 A1, which is hereby incorporated into the present application by reference.

The propylene oxide can also be treated with suitable adsorbents, as is taught, for example, in EP 0 675 119 A2 or GB 1,035,866 A, which are both hereby incorporated into the present application by reference.

Suitable methods for the purification of PO which has been prepared by means of the SM/PO process are, for example, treatment with ion exchange resins, as disclosed in US 5,107,007 A, or extractive distillation, as disclosed, for example, in US 3,909,366 A, which are both hereby incorporated into the present application by reference.

For the purification of PO which has been prepared by means of the MTBE/PO process, a suitable method is, for example, extractive distillation, as disclosed in US 5,006,206 A, which is hereby incorporated into the present application by reference, or in US 5,133,839 A.

For the purification of PO which has been prepared by means of the epichlorohydrin process, a suitable method is, for example, fractional distillation with subsequent stripping of the side stream, as is proposed in US 3,398,062 A, which is hereby incorporated into the present application by reference.

For the purification of PO which has been prepared by means of the HPPO process, a suitable method is, for example, extractive distillation, as disclosed in WO 2004/092150 A1, which is hereby incorporated into the present application by reference.

The invention will be further illustrated by the nonlimiting examples below.

### **Examples**

#### Test method A

The catalyst testing was carried out in the following way: the starter polyol used was a polypropylene glycol with molar mass 400 which was prepared with KOH catalysis and from which the catalyst was removed by means of phosphoric acid precipitation, filtration and subsequent treatment with Ambosol<sup>®</sup>, a commercially available magnesium silicate. The DMC catalyst used was prepared as in EP 0 862 947 and employed as a solid.

128 g of starter and 0.03 g of catalyst were introduced as initial charge into a 250 ml reactor with magnetic stirrer, heated to 100°C and evacuated for 2 h at < 5 mbar. The vacuum was then lifted with nitrogen, the reaction mixture was heated to 130°C and the nitrogen pressure was adjusted to 1.5 bar. The troublesome substance to be tested was introduced into the reactor through a septum using a syringe. It was then flushed with 1 to 2 g of starter which was inertized beforehand with nitrogen.

In the case of formaldehyde, the procedure differed as follows: solid paraformaldehyde was charged directly to the reactor following evacuation and with nitrogen blanketing. After closing the reactor, it was heated to 200°C in order

to depolymerize the paraformaldehyde. A temperature of 130°C was then established as above.

At 130°C, 72 g of propylene oxide (PO) were then introduced into the reactor over the course of 2 min. The resulting pressure and temperature changes were recorded for two hours. From the maxima, the initiation time, i.e. the time period between adding PO and the onset of the alkoxylation reaction, was determined. Moreover, the yield of the resulting product was determined.

#### Test method B

Into a 300 ml autoclave, 40 g of polypropylene glycol with OH number 100 and functionality 1.98 and 70 ppm of DMC catalyst (based on the final amount of the polyol) were introduced as initial charge. At 130°C and with stirring, flushing with nitrogen was carried out twice at 5 bar. 7 bar of nitrogen was then applied for 20 min. The system was then held under vacuum for 25 min. In phase 1 of the test, with stirring, 4.8 ml of in total 151 g of PO were introduced at 2.5 ml/min at 130°C and max. 4 bar. Phase 2 followed when the exothermic initiation reaction had subsided and the pressure had dropped below 0.7 bar; if not, phase 1 was repeated. In phase 2, at 100°C and initially vacuum, the remaining PO was added over the course of 22 min, the introduction rate being increased in stages from 0 to 5.0 ml/min; during this, a pressure from 1.9 to 5.5 bar was established. A post-reaction time of 60 min then followed. The mixture was worked-up as follows: holding at vacuum for 25 min at 100°C, then cooling to 25°C and flushing with 2 bar of nitrogen. OH number and viscosity were determined for the product (theoretical OH number: 22 mg KOH/g).

The viscosity of the resulting polyetherols was determined according to DIN 53019 using a rotary viscometer of the Couette type, at the temperature stated in Table 1 and a shear stress  $\tau$  (definition in DIN 1342) of 1000 Pa. For this, the viscosity was measured both at a speed which produced a shear stress which was somewhat less than 1000 Pa, and also at a speed at which a shear stress resulted which was somewhat greater than 1000 Pa. The two measured viscosities (at  $\tau < 1000$  Pa and  $\tau > 1000$  Pa) were used to determine the viscosity at  $\tau = 1000$  Pa by linear interpolation.

The results are summarized in Tables 1/1 to 1/3. The theoretical yield was 200 g.

As can be seen, in the presence of the tested troublesome substances, the initiation time is in some cases significantly extended, the extent of the extension depending on the concentration of the troublesome substance in question. In some cases, no reasonable yield is achieved and deviations in the product properties result.



Table 1/1: Results of Test A, (initiation time,  $T_{max}$  and  $P_{max}$ ) and Test B (yield, OH number and viscosity)

Troublesome substance	Conc. [ppm]	Equivalent concentration [ppm]	Initiation time [min]	$T_{max}$ [°C]	$P_{max}$ [bar]	Yield [g]	OH number [mg KOH/g]	Viscosity [mPa*s, 25°C]
Without	0		5	280	11.2	196	21.6	2626
Acetaldehyde	1190	calc. propionaldehyde: 1569	11	269	11.7	196	21.9	3192
Acetaldehyde	466	calc. propionaldehyde: 614	7	264	11.1	196	22.1	2803
Acetic acid	600		29	277	12.4	196	21.9	5732
Acetic acid	169		15	273	14.5	198	21.5	3635
Acetone	980		12	277	10.1	198	21.5	3297
Acetone	50		5	265	15	195	21.5	2626
Allyl alcohol	1705		11	248	10.3	190	22.1	4520
Allyl alcohol	230		4	207	13.5	190	21.5	2542
1-Bromo-2-propanol	61	calc. Br: 35	22	250	10.5	196	22.2	4565
1-Bromo-2-propanol	24	calc. Br: 14	5	279	11	196	21.9	2626
4-Chloro-1-butanol	370	calc. Cl: 121	did not start	130	7.5	-	-	-
4-Chloro-1-butanol	71	calc. Cl: 23	did not start	130	7.1	-	-	-

Table 1/2

Troublesome substance	Conc. [ppm]	Equivalent concentration [ppm]	Initiation time [min]	T <sub>max</sub> [°C]	P <sub>max</sub> [bar]	Yield [g]	OH number [mg KOH/g]	Viscosity [mPa*s, 25 °C]
4-Chloro-1-butanol	26	calc. Cl: 8.5	did not start	132	7.1	-	-	-
1-Chloro-1-propene	483	calc. Cl: 224	13	284	14.3	198	21.4	3406
1-Chloro-1-propene	50	calc. Cl: 23	7	259	13.4	198	21.5	2803
2-Chloro-1-propene	550	calc. Cl: 255	19	266	11.9	196	21.9	4140
3-Chloro-1-propene	1030	calc. Cl: 477	33	217	11.3	197	21.7	6528
3-Chloro-1-propene	565	calc. Cl: 262	4	270	11.5	195	21.4	2542
2-Chloropropane	2425	calc. Cl: 1095	10	220	11.8	198	21.9	3090
2-Chloropropane	50	calc. Cl: 23	8	257	11.3	196	21.4	3010
1-Chloro-2-propanol	92	calc. Cl: 35	49	152	8	197	22.5	10983
1-Chloro-2-propanol	50	calc. Cl: 19	19	256	10.6	196	22.1	4140
3-Chloro-1-propanol	633	calc. Cl: 237	8	254	11	196	21.7	2895

Table 1/3

Troublesome substance	Conc. [ppm]	Equivalent concentration [ppm]	Initiation time [min]	T <sub>max</sub> [°C]	P <sub>max</sub> [bar]	Yield [g]	OH number [mg KOH/g]	Viscosity [mPa*s, 25 °C]
3-Chloro-1-propanol	293	calc. Cl: 110	5	270	10.6	196	21.9	2626
Allyl chloride	298	calc. Cl: 138	37	196	8.4	195	21.9	7435
Allyl chloride	117	calc. Cl: 52	13	271	14.2	197	21.7	3685
Cyclohexane	1005		9	275	10	196	21.5	2991
Cyclohexane	50		4	264	10.1	196	21.7	2542
Formaldehyde	25	calc. propionaldehyde: 48	41	214	9.9	197	22.3	8467
Formaldehyde	10	calc. propionaldehyde: 19	25	251	12.6	197	21.8	5032
Water	80		16	238	11.4	197	21.9	3755
Water	68		14	244	10	196	21.6	3519
Water	50		8	268	10.9	198	21.6	3120

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**Claims**

1. A process for preparing polyols by means of the addition reaction of alkylene oxides onto H-functional starter molecules in the presence of DMC catalysts, 10 wherein, during the alkoxylation, propylene oxide is used in which the sum of the contents of organohalogen compounds, determined in accordance with DIN 51408-2 and EN ISO 10304, is not more than 150 ppm; in which the sum of 15 the contents of aldehydes and ketones, determined in accordance with VDI 3862 Part 2, is not more than 300 ppm; whose content of allyl alcohol determined by means of gas chromatography analogously to DIN 51405, is not more than 8500 ppm; and whose content of hydrocarbons, the content of 20 which is determined by means of gas chromatography analogously to DIN 51405 and is expressed as the sum value, is not more than 500 ppm.

2. The process according to claim 1, in which the content 25 of organohalogen compounds in the propylene oxide is not more than 60 ppm.

3. The process according to claim 1 or 2, in which the content of aldehydes and ketones in the propylene oxide is 30 not more than 120 ppm.

4. The process according to any one of claims 1 to 3, in which the content of allyl alcohol in the propylene oxide is not more than 3400 ppm.

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5. The process according to any one of claims 1 to 4, in which the content of hydrocarbons in the propylene oxide is not more than 200 ppm.

5 6. The process according to any one of claims 1 to 5, in which, during the alkoxylation, propylene oxide is used whose content of water, determined by Karl-Fischer titration in accordance with DIN EN 13267, is not more than 500 ppm.

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7. The process according to claim 6, in which the content of water in the propylene oxide is not more than 200 ppm.

8. The process according to any one of claims 1 to 7, in which, during the alkoxylation, propylene oxide is used whose content of acid, expressed by the value determined by means of acid-base titration in accordance with DIN EN 62021-2 or DIN 12634 and calculated for acetic acid, is not more than 100 ppm.

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9. The process according to claim 8, in which the content of acid in the propylene oxide is not more than 40 ppm.

10. The process according to any one of claims 1 to 9, in which, during the alkoxylation, propylene oxide is used whose content of ethylene oxide, determined by gas chromatography analogously to DIN 51405, is not more than 100 ppm.

30 11. A polyol obtainable by a process according to any one of claims 1 to 10.

12. The use of the polyols according to claim 11 for producing polyurethanes.

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13. The use of the polyols according to claim 11 for producing polyurethane foams.

14. The use of the polyols according to claim 11 for  
5 producing polyurethanes for coatings, adhesives, sealing compositions or elastomers (CASE).

15. The use of propylene oxide whose content of organo-  
halogen compounds, determined in accordance with  
10 DIN 51408-2, is not more than 150 ppm; whose content of aldehydes and ketones, determined in accordance with  
DIN 51373, is not more than 300 ppm; whose content of allyl  
alcohol, determined by means of gas chromatography  
analogously to DIN 51405, is not more than 8500 ppm; and  
15 whose content of hydrocarbons, whose content is determined  
by means of gas chromatography analogously to DIN 51405 and  
is expressed as the sum value, is not more than 500 ppm for  
the alkoxylation of H-functional starter molecules in the  
presence of DMC catalysts.

## INTERNATIONAL SEARCH REPORT

International application No  
PCT/EP2010/007641

A. CLASSIFICATION OF SUBJECT MATTER INV. C08G65/26 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) 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) EPO-Internal		
C. DOCUMENTS CONSIDERED TO BE RELEVANT		
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	WO 2005/040248 A1 (SHELL INT RESEARCH [NL]; BECKERS JOHANNES GERHARDUS JOS [NL]; ELEVELD) 6 May 2005 (2005-05-06) claims, examples; page 1 - page 3 in particular comparative examples 1 and 2 -----	1-15
X	WO 2004/076528 A1 (SHELL INT RESEARCH [NL]; BECKERS JOHANNES GERHARDUS JOS [NL]; DU CAUZE) 10 September 2004 (2004-09-10) page 1 - page 3 page 1 line 26 claims, examples in particular comparative examples 1 and 2 ----- -/--	1-15
<input checked="" type="checkbox"/> Further documents are listed in the continuation of Box C. <input checked="" type="checkbox"/> 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 document but published on or after the international filing date "L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified) "O" document referring to an oral disclosure, use, exhibition or other means "P" document published prior to the international filing date but later than the priority date claimed		"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. "&" document member of the same patent family
Date of the actual completion of the international search  14 March 2011		Date of mailing of the international search report  22/03/2011
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  Kositza, Matthias

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International application No  
PCT/EP2010/007641

C(Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT		
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