



EUROPEAN PATENT APPLICATION

Application number : **91311636.4**

Int. Cl.⁵ : **C11D 1/00, C11D 3/20**

Date of filing : **13.12.91**

Priority : **18.12.90 GB 9027414**

Date of publication of application :
24.06.92 Bulletin 92/26

Designated Contracting States :
CH DE ES FR GB IT LI NL SE

Applicant : **UNILEVER PLC**
Unilever House Blackfriars P.O. Box 68
London EC4P 4BQ (GB)
GB

Applicant : **Unilever N.V.**
Burgemeester s'Jacobplein 1
NL-3015 CA Rotterdam (NL)
CH DE ES FR IT LI NL SE

Inventor : **Falou, Mohamad Sami**
Unilever Res.Port Sunlight Lab., Quarry Road
East
Bebington, Wirral, Merseyside L63 3JW (GB)
Inventor : **Gough, Anthony David**
Unilever Res.Port Sunlight Lab., Quarry Road
East
Bebington, Wirral, Merseyside L63 3JW (GB)
Inventor : **Hull, Michael**
Unilever Res.Port Sunlight Lab., Quarry Road
East
Bebington, Wirral, Merseyside L63 3JW (GB)

Representative : **Fransella, Mary Evelyn et al**
Unilever PLC Patents Division P.O. Box 68
Unilever House
London EC4P 4BQ (GB)

Detergent compositions.

Detergent compositions for washing fabrics contain a mixture of alkylpolyglycoside surfactant and alkane 1,2-diol. The present of minority amounts of diol gives synergy with the alkylpolyglycoside, manifested by enhanced removal of oily soil.

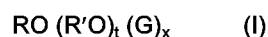
Field of the Invention

This invention relates to detergent compositions, particularly but not exclusively to built detergent compositions for washing fabrics.

Background of the Invention

Detergent compositions traditionally contain one or more detergent active materials in addition to various other ingredients such as detergency builders, bleaches, fluorescers, perfumes etc. Notable applications of detergent compositions are to clean fabrics, usually by washing portable fabric items in a bowl or in a washing machine, to clean crockery and cooking utensils, again by washing in a bowl (hand dishwashing), and to clean hard surfaces such as glass, glazed surfaces, plastics, metals and enamels. A number of classes of surfactant materials have been used as detergent active materials, including anionic and nonionic materials.

One known category of nonionic surfactants are compounds which are often known as alkylpolyglycosides. These are of the general formula



in which R is an organic hydrophobic residue, R'O is an alkoxy group which may be absent because t can be zero, and G is a saccharide residue and x is at least unity. A more detailed definition is set out hereinafter.

Objective of the Invention

We have now found that a combination of alkylpolyglycoside with certain diols which are themselves nonionic surfactants provides unexpected advantages provided their weight ratio lies within certain limits.

Such combinations have been found to give a synergistic benefit of enhanced oily/fatty soil detergency.

The Invention

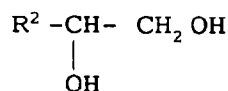
According to the present invention there is provided a detergent composition containing

i) an alkylpolyglycoside of the general formula



in which R is an organic hydrophobic residue containing 10 to 20 carbon atoms, R' contains 2 to 4 carbon atoms, G is an saccharide residue containing 5 or 6 carbon atoms, t is in the range 0 to 25 and x is in the range from 1 to 10;

ii) a 1,2-diol of the general formula



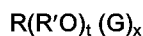
wherein R² is a saturated or unsaturated organic hydrophobic residue containing 8 to 16 carbon atoms; the weight ratio of the surfactants (i) and (ii) lying in the range from 40:1 to 7:3.

The weight ratio of the two surfactants, may lie in a range from 40:1 to 3:1 or 4:1, or a still narrower range from 20:1 to 6:1 or even 15:1 to 8:1. The weight ratio for optimum synergy will vary depending on the surfactants used and any other surfactants present. It can be determined by experiment.

The invention also provides a method of washing which comprises contacting fabrics, or a surface to be cleaned, with a composition according to this invention or a wash liquor obtainable by adding the composition to water, notably in an amount ranging from 0.5 to 50 grams of composition per litre of water.

The alkylpolyglycoside

In the general formula



the hydrophobic group R is preferably aliphatic, either saturated or unsaturated, notably straight or branched alkyl, alkenyl, hydroxyalkyl or hydroxyalkenyl. However, it may include an aryl group for example alkyl-aryl, alkenyl-aryl and hydroxyalkyl-aryl. Particularly preferred is that R is alkyl or alkenyl of 8 to 16 carbon atoms.

The value of t in the general formula above is preferably zero, so that the -(RO)_t- unit of the general formula is absent. In that case the general formula becomes



If t is non-zero it is preferred that R'O is an ethylene oxide residue. Other likely possibilities are propylene oxide and glycerol residues. If the parameter t is non-zero so that R'O is present, the value of t (which may be an average value) will preferably lie in the range from 0.5 to 10.

The group G is typically derived from fructose, glucose, mannose, galactose, talose, gulose, allose, altrose, idose, arabinose, xylose, lyxose and/or ribose. Preferably, the G is provided substantially exclusively by glucose units.

The value x, which is an average, is usually termed the degree of polymerization. Desirably x varies between 1 and 8. Values of x may lie between 1 and 3, especially 1 and 1.8.

Polyglycosides of particular interest have x in the narrow range from 1 or 1.2 up to 1.4 or especially 1.3. If x exceeds 1.3 it preferably lies in the range 1.3 or 1.4 to 1.8.

When x lies in the range 1 to 1.4 it is preferred that R is C₈ to C₁₄ alkyl or alkenyl. The even narrower range of C₈ to C₁₂ may be used.

The Diol (ii)

Alkane diols for use in this invention are generally hydrophobic in character. This is manifested by formation of a turbid dispersion rather than an isotropic solution when placed, alone, in deionized water at a surfactant concentration of 1% or more by weight.

In the diol, the hydrophobic residue R² will generally be alkyl or alkenyl of 8 to 16 carbon atoms, preferably 8 to 12. 1,2-Alkane diols are known compounds which can be prepared by standard methods for the preparation of diols. Notably, they can be made from terminal olefins by epoxidation and hydrolysis.

Other possibilities for the hydrophobic residue R include cycloaliphatic groups and aromatic-aliphatic groups such as phenylethyl or dimethylphenyl.

Detergent compositions of the invention may contain further surfactants, outside the definitions stated for (i) and (ii). The nature and amount of any additional surfactant may affect the synergistic action of the specified surfactants (i) and (ii). Detergency should therefore be checked if additional surfactant is included. In particular, additional non-soap anionic surfactant may be avoided or used only in small amount. The amount of any additional surfactant will frequently be less than 50% by weight, and perhaps not over 40%, 25% or even 10% by weight of the overall surfactant mixture.

Additional surfactant, if present, may be anionic, nonionic or amphoteric. Cationic surfactant is possible if anionic surfactant is absent.

In particular, nonionic surfactant with an HLB value greater than 10.5 may be present. This may for instance be ethoxylated fatty alcohol which affects the proportions giving synergy but does not greatly harm the synergy. Another surfactant which may be included is soap. If included, soap may be derived from saturated fatty acid or a mixture of saturated and unsaturated fatty acids. Soap may be an alkali metal salt, especially a sodium salt of fatty acid. It may be an ammonium or substituted ammonium salt, but this is less likely.

Compositions of this invention will generally contain a surfactant mixture comprising (i) the specified alkyl-polyglycoside (ii) the specified nonionic surfactant and (iii) any other surfactant(s), in a total amount which is from 1 to 60% by weight of the composition. Preferred amounts are 2 to 45%, better 5 to 40% or 35%. The total of the specified surfactant (i) and (ii) may itself be at least 2% or at least 5% of the overall composition.

The compositions of the invention may contain an electrolyte, for instance present in such an amount to give a concentration of at least 0.01 molar, when the composition is added to water at a concentration of 1 g/litre. Electrolyte concentration may possibly be higher such as at least 0.05 or 0.1 molar especially if the composition is of solid form; liquid compositions generally limit electrolyte for the sake of stability. 1 g/litre is approximately the lowest level at which detergent compositions for fabric washing are used in usual practice. More usual is usage at a level of 4 to 50 g/litre. The amount of electrolyte may be such as to achieve an electrolyte concentration of 0.01 molar, most preferably at least 0.1 molar, when the composition is added to water at a concentration of 4 g/litre.

If the composition of the invention is intended as a fabric washing composition it will generally contain detergency builder in an amount from 7 to 70% by weight of the composition.

If it is in solid form, the composition is likely to contain at least 10 or 15% of builder.

It is desirable that the compositions according to the invention be approximately neutral or at least slightly alkaline, that is when the composition is dissolved in an amount to give surfactant concentration of 1 g/l in distilled water at 25°C the pH should desirably be at least 7.5. For solid compositions the pH will usually be greater, such as at least 9. To achieve the required pH, the compositions may include a water-soluble alkaline salt. This salt may be a detergency builder (as described in more detail below) or a non-building alkaline material.

When the compositions of the invention contain a detergency builder material, this may be any material

capable of reducing the level of free calcium ions in the wash liquor and will preferably provide the compositions with other beneficial properties such as the generation of an alkaline pH and the suspension of soil removed from the fabric.

5 Examples of phosphorus-containing inorganic detergency builders, when present, include the water-soluble salts, especially alkali metal pyrophosphates, orthophosphates, polyphosphates and phosphonates. Specific examples of inorganic phosphate builders include sodium and potassium tripolyphosphates, orthophosphates and hexametaphosphates.

10 Examples of non-phosphorus-containing inorganic detergency builders, when present, include water-soluble alkali metal carbonates, bicarbonates, silicates and crystalline and amorphous aluminosilicates. Specific examples include sodium carbonate (with or without calcite seeds), potassium carbonate (with or without calcite seeds), sodium and potassium bicarbonates and silicates.

15 Examples of organic detergency builders, when present include the alkali metal, ammonium and substituted ammonium polyacetates, carboxylates, polycarboxylates, polyacetyl carboxylates and polyhydroxysulphonates. Specific examples include sodium, potassium, lithium, ammonium and substituted ammonium salts of ethylenediaminetetraacetic acid, nitrilotriacetic acid, oxydisuccinic acid, melitic acid, benzene polycarboxylic acids and citric acid. Further possibilities are tartrate monosuccinates, tartrate disuccinates, dipicolinic acid, cheledamic acid, carboxymethyl oxysuccinate and hydroxy ethyl imino diacetic acid.

20 Apart from the ingredients already mentioned, a number of optional ingredients may also be present. Examples of other ingredients which may be present in the composition are polymers containing carboxylic or sulphononic acid groups in acid form or wholly or partially neutralised to sodium or potassium salts, the sodium salts being preferred. Preferred polymers are homopolymers and copolymers of acrylic acid and/or maleic acid or maleic anhydride. Of especial interest are polyacrylates, polyalpha-hydroxy acrylates, acrylic/maleic acid copolymers, and acrylic phosphinates. Other polymers which are especially preferred for use in liquid detergent compositions are deflocculating polymers such as for example disclosed in EP 346995.

25 The molecular weights of homopolymers and copolymers are generally 1000 to 150,000, preferably 1500 to 100,000. The amount of any polymer may lie in the range from 0.5 to 5% by weight of the composition. Other suitable polymeric materials are cellulose ethers such as carboxy methyl cellulose, methyl cellulose, hydroxy alkyl celluloses, and mixed ethers, such as methyl hydroxy ethyl cellulose, methyl hydroxy propyl cellulose, and methyl carboxy methyl cellulose. Mixtures of different cellulose ethers, particularly mixtures of carboxy methyl cellulose and methyl cellulose, are suitable. Polyethylene glycol of molecular weight from 400 to 50,000, preferably from 1000 to 10,000, and copolymers of polyethylene oxide with polypropylene oxide are suitable as also are copolymers of polyacrylate with polyethylene glycol. Polyvinyl pyrrolidone of molecular weight of 10,000 to 60,000 preferably of 30,000 to 50,000 and copolymers of polyvinyl pyrrolidone with other poly pyrrolidones are suitable. Polyacrylic phosphinates and related copolymers of molecular weight 1000 to 100,000, 35 in particular 3000 to 30,000 are also suitable.

Further examples of other ingredients which may be present in the composition include fabric softening agents such as fatty amines, fabric softening clay materials, lather boosters such as alkanolamides, particularly the monoethanolamides derived from palm kernel fatty acids and coconut fatty acids, lather depressants, oxygen-releasing bleaching agents such as sodium perborate and sodium percarbonate, typically accompanied 40 by peracid bleach precursors, organic peracids, chlorine-releasing bleaching agents such as trichloroisocyanuric acid, inorganic salts such as sodium sulphate, and, usually present in very minor amounts, fluorescent agents, perfumes including deodorant perfumes, enzymes such as cellulases, proteases, lipases and amylases, germicides and colourants.

45 The detergent compositions according to the invention may be in any suitable form including powders, bars, liquids and pastes. For example suitable liquid compositions may be non-aqueous or aqueous, the latter being either isotropic or lamellar structured. The compositions may be prepared by a number of different methods according to their physical form. In the case of granular products they may be prepared by dry-mixing, coagglomeration, spray-drying from an aqueous slurry or any combination of these methods. One preferred physical form is a granule incorporating a detergency builder salt. This may be prepared by conventional granulation techniques or spray drying. 50

Another preferred physical form is a lamellar structured aqueous liquid. Structuring a liquid by means of surfactant is well known and may be utilised to provide consumer-preferred flow properties, and/or turbid appearance. Also many liquids in which the surfactant mixture provides structure are capable of suspending particulate solids such as detergency builders and abrasives. For such forms, alkyl polyglycoside which are particularly suitable have an HLB of at least 12.0 and in its formula 55



t is zero or 1 to 3, preferably zero, while

x is 1 to 3, especially 1 to 1.8.

The aqueous continuous phase will usually contain some dissolved electrolyte. Electrolyte may be dissolved only in the aqueous continuous phase or may also be present as suspended solid particles. Particles of solid materials which are insoluble in the aqueous phase may be suspended alternatively or in addition to any solid electrolyte particles.

Although structured liquids require some electrolyte to be present in the continuous phase, the amount which is present generally has to be limited for the sake of stability. When the present invention takes the form of a structured liquid, an advantage is that the structuring conferred by the surfactant mixture of the invention will tolerate a substantial amount of electrolyte.

Three common product forms which are of the structured liquid type are liquids for heavy duty fabrics washing, liquid abrasives and general purpose cleaners. In the first class, the suspended solid can comprise suspended solids which are substantially the same as the dissolved electrolyte, being an excess of same beyond the solubility limit. This solid is usually present as a detergency builder, i.e. to counteract the effects of calcium ion water hardness in the wash. In the second class, the suspended solid usually comprises a particulate abrasive, insoluble in the system. In that case the electrolyte, present to contribute to the structuring of the active material in the dispersed phase, is generally different from the abrasive compounds. In certain cases, the abrasive can however comprise partially soluble salts which dissolve when the product is diluted. In the third class, the structure is usually used for thickening the product to give consumer-preferred flow properties, and sometimes to suspend pigment particles.

In the Examples which follow, the parts and percentages are by weight. The sole drawing is a triangular plot derived from one example.

Example 1

Aqueous wash liquors were prepared containing the following materials in deionised water.

Alkyl polyglycoside)	
)	1 g/litre
Dodecane 1,2-diol)	
Sodium metaborate		0.05 molar

These quantities would be typical of using 6g/litre of a detergent product containing 16.7% by weight surfactant. The wash liquors had a pH of about 10 resulting from the presence of the metaborate.

The alkylpolyglycoside was APG 300 from Horizon Chemical Co. This was of the formula



where R is a 9 to 11 carbon alkyl chain, G is glucose and x has an average value of 1.4. It had an HLB value of approximately 11 to 12. The dodecane 1,2-diol is available from Aldrich Chemical Co.

Wash liquors were prepared with various ratios of the two surfactants and used to wash polyester test cloths soiled with radiolabelled triolein. Washing was carried out at 40°C for 20 minutes in a Tergotometer.

The removal of triolein was determined and the results were:

	Ratio	% Triolein removal
	APG 300/1,2-diol	
5		
	100/0	58.9
	95/5	60.2
10	90/10	58.5
	80/20	41.2
15	60/40	4.5
	40/60	2.2
	20/80	1.7
20	0/100	1.7

Example 2

25 Example 1 was repeated using a different alkylpolyglycoside. APG 500 from Horizon was used. This has the formula



where R is C₁₂ and C₁₃ alkyl, G is glucose and x is 1.4. It had an HLB value of approximately 11.5 - 12.5. Results were:

	Ratio	% Triolein removal
	APG 500:1,2-diol	
30		
	100:0	45.3
	90:10	54.1
40	80:20	37.1
	60:40	6.2
45	40:60	2.3
	20:80	1.8
	0:100	1.7

50 It can be seen from the Examples 1 and 2 above that there is a mixture of surfactants which gives better triolein removal than either individual surfactant. The proportions giving optimum synergy can be found by experiments such as these.

55 Example 3

Compositions were prepared having as a general formulation:

	% by weight
Detergent active	13
Zeolite	24
Sodium silicate	0.8
Sokolan CP5	4
Sodium carbonate (anhydrous)	12
Sodium carboxymethyl cellulose	0.5
Sodium metaborate	11
Water	balance to 100%

Sokolan CP5 is an acrylic/maleic copolymer available from BASF.

The detergent active was provided by various proportions of an alkylpolyglycoside and dodecane 1,2-diol. The alkylpolyglycoside was APG 550 from Horizon. This is of the general formula



where R is a 12 and 13 carbon atom alkyl chain, G is glucose and x has an average value of 1.8. It has an HLB value of approximately 12 to 13.

The compositions were added to 24°FH water at a concentration of 6g/litre.

The resulting wash liquors had a pH of 10 and were used to wash polyester test cloths as in Examples 1 and 2.

Results were:

Ratio	% Triolein removal
APG 550:1,2-diol	
100:0	43.5
92:8	58.0
85:15	32.2
69:31	5.9
54:46	2.6

Example 4

The procedure of Example 3 was repeated using mixtures of APG 550, Dodecane 1,2-diol and Synperonic A7 which is a C₁₃-C₁₅ alcohol ethoxylated with an average of seven ethylene oxide residues. It has an HLB value of 11.7.

The results are set out in the following table in which the amounts of APG 550, Dodecane 1,2-diol and Synperonic A7 are expressed as percentages of the total quantity of all three (which was a constant 13% of the overall composition). The results are also plotted as a triangular diagram which appears as the sole drawing.

	APG 550	Synperonic A7	C ₁₂ 1,2 Diol	% Triolein Removal
5	0	100	0	68.9
	15.4	84.6	0	66.9
	30.8	69.2	0	64.2
10	50.0	50.0	0	62.5
	69.2	30.8	0	57.4
	100	0	0	43.5
15	80.8	11.5	7.7	63.3
	92.3	0	7.7	58.0
20	0	84.6	15.4	74.5
	23.1	61.5	15.4	68.0
	53.8	30.8	15.4	63.8
25	69.2	15.4	15.4	58.5
	84.6	0	15.4	32.2
30	0	69.2	30.8	62.7
	15.4	53.8	30.8	59.2
	23.1	46.1	30.8	55.0
35	38.4	30.8	30.8	47.8
	69.2	0	30.8	5.9
	0	53.8	46.2	13.6
40	23.1	30.8	46.2	11.3
	38.4	15.4	46.2	8.7
45	53.8	0	46.2	2.6

It can be seen from this triangular diagram that as the mixture contains increasing quantities of Synperonic A7, there is still a ratio of APG 550 : diol which gives greater triolein removal than either of them alone, with the same quantity of Synperonic A7. This can be seen in particular from the results with 30.8% Synperonic A7 present.

Example 5

Compositions were prepared having as a general formulation:

		% by weight
	Detergent active	17
5	Zeolite	32
	Sodium silicate	0.8
	Sokolan CP5	4.0
10	Sodium carbonate (anhydrous)	14.5
	Sodium metaborate	16.5

15 The detergent active was provided by various proportions of alkylpolyglycoside and dodecane 1,2-diol. The alkylpolyglycoside was APG 600 from Horizon (also available as Plantaren APG 600 from Henkel). This is of the general formula

$RO(G)_x$

20 where R is a coconut alkyl chain (i.e. principally C_{12} and C_{14}), G is glucose and x has an average value of 1.4. It has an HLB value of approximately 12.

The compositions were added to 24° FH water at a concentration of 5g/litre and used to wash polyester test cloths by the procedure as in Examples 1 and 2.

Results were:

25	Ratio	% Triolein removal
	APG 600:1,2-diol	
30	100:0	60.3
	93:7	66.7
35	86:14	70.0
	79:21	64.1
	72:28	57.1
40	51:49	7.8
	30:70	1.7

45 The synergy over a range of ratios is apparent.

Example 6

50 Example 5 was repeated using ternary mixtures of APG 600, dodecane 1,2-diol, and coconut alkyl sulphate (PAS).

Results were:

55

	Ratio:			% Triolein removal
	APG 600 : 1,2-diol : PAS			
5	70	:	70 : 30	3.0
	75	:	0 : 25	49.2
10	61	:	14 : 25	60.4
	69	:	14 : 17	63.3
15	63	:	21 : 16	63.6
	45	:	35 : 19	48.5

It is thus shown that there is a synergistic improvement with certain ratios of APG and diol when alkyl sulphate is also present as compared with just APG or diol and a similar amount of the coconut alkyl sulphate.

Claims

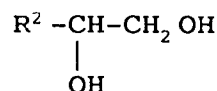
1. A detergent composition containing

i) an alkylpolyglycoside of the general formula



in which R is an organic hydrophobic residue containing 10 to 20 carbon atoms, R' contains 2 to 4 carbon atoms, G is a saccharide residue containing 5 or 6 carbon atoms, t is in the range 0 to 25 and x is in the range from 1 to 10;

ii) a 1,2-diol of the general formula



where R² is a saturated or unsaturated organic hydrophobic residue containing 8 to 16 carbon atoms;

the weight ratio of the surfactants (i) and (ii) lying in the range from 40:1 to 7:3.

2. A detergent composition according to claim 1 wherein the weight ratio of the surfactants (i) and (ii) lies in the range from 40:1 to 4:1.

3. A detergent composition according to claim 1 wherein the weight ratio of the surfactants (i) and (ii) lies in the range from 20:1 to 6:1.

4. A detergent composition according to any one of the preceding claims wherein the alkylpolyglycoside has a value of t which is zero so that it is of the general formula



5. A detergent composition according to any one of the preceding claims wherein the alkylpolyglycoside (i) has an average value of x in the range from 1 to 1.8.

6. A detergent composition according to any one of claims 1 to 4 wherein the alkylpolyglycoside (i) has an average value of x in the range from 1 to 1.4.

7. A detergent composition according to any one of claims 1 to 4 wherein the alkylpolyglycoside (i) has an

average value of x in the range from 1.3 to 1.8.

- 5 8. A detergent composition according to any one of the preceding claims wherein the organic hydrophobic residue R of the diol (ii) is an alkyl or alkenyl group of 8 to 16 carbon atoms.
9. A detergent composition according to any one of the preceding claims comprising 7 to 70% by weight of detergency builder.
- 10 10. A detergent composition according to any one of the preceding claims comprising 1 to 60% by weight of a surfactant mixture containing the said alkylpolyglycoside (i) and the said nonionic surfactant (ii) in amounts which total 60 to 100% by weight of the surfactant mixture, together with other ingredients and/or water.
11. A detergent composition according to claim 10 wherein the alkylpolyglycoside (i) and the alkane diol (ii) 15 constitute 75 to 100% by weight of the surfactant mixture.
12. A detergent composition according to claim 10 wherein the alkylpolyglycoside (i) and the alkane diol (ii) constitute 90 to 100% by weight of the surfactant mixture.
- 20 13. A method of cleaning which comprises contacting fabrics or other inanimate surface to be cleaned with a composition according to any one of the preceding claims or a wash liquor comprising water and a composition according to any one of the preceding claims added to the water in a quantity lying in a range from 0.5 to 50 grams per litre of water.

25

30

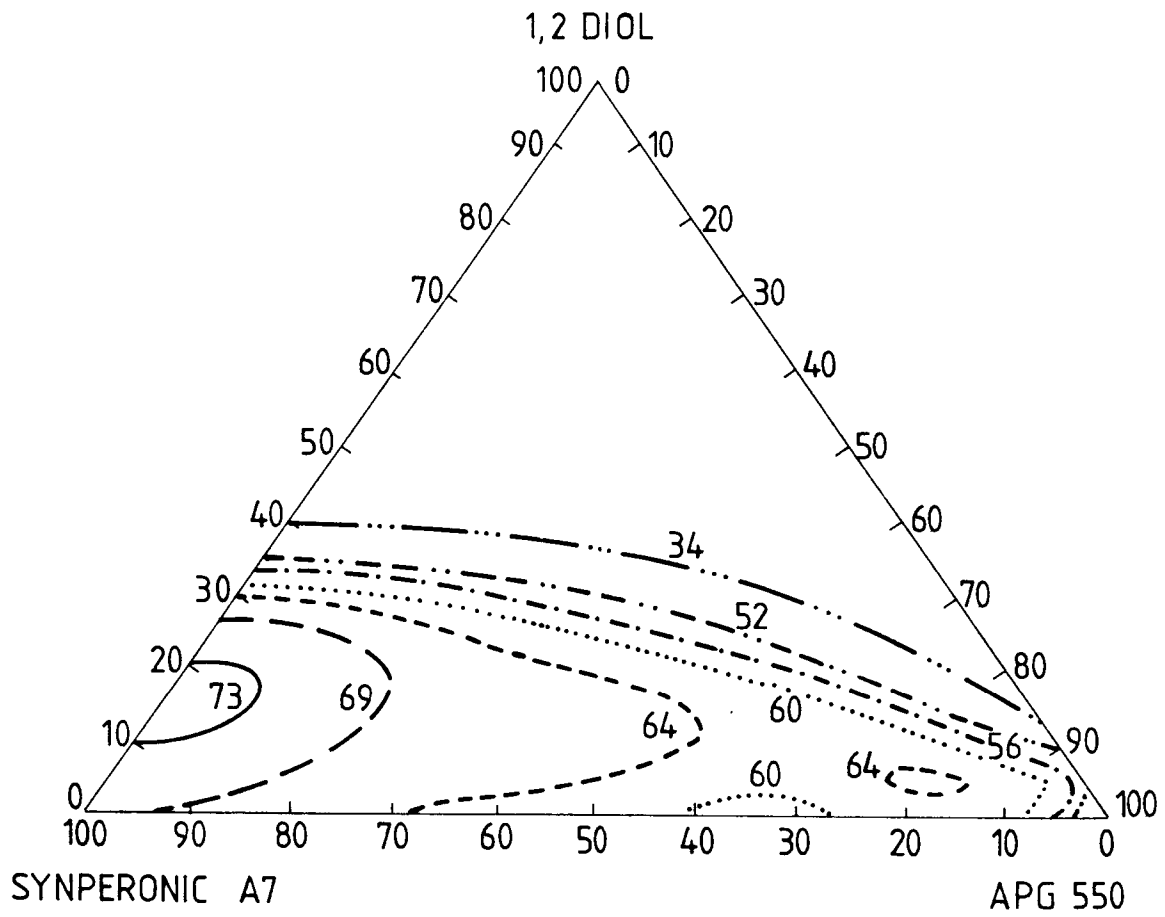
35

40

45

50

55



% REMOVAL

- 73.0
- - - 68.7
- - - 64.4
- 60.1
- · - · 55.8
- · - · 51.5
- 34.25



European Patent
Office

EUROPEAN SEARCH REPORT

Application Number

DOCUMENTS CONSIDERED TO BE RELEVANT			EP 91311636.4
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int. Cl.5)
A	US - A - 4 483 780 (LIENADO) * Claim 1 * --	1, 4, 5, 6	C 11 D 1/00 C 11 D 3/20
A	US - A - 4 536 319 (PAYNE) * Claim 1 * -----	1, 4, 5, 6	
			TECHNICAL FIELDS SEARCHED (Int. Cl.5)
			C 11 D
The present search report has been drawn up for all claims			
Place of search VIENNA		Date of completion of the search 04-03-1992	Examiner SEIRAFI
<p>CATEGORY OF CITED DOCUMENTS</p> <p>X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure P : intermediate document</p> <p>T : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons & : member of the same patent family, corresponding document</p>			

EPO FORM 1503 03.82 (PC401)