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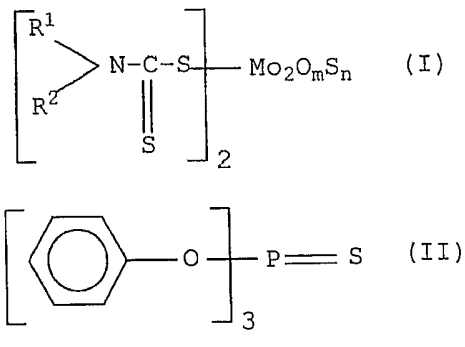
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WO 2004/113480 A1

(54) Title: UREA GREASE COMPOSITION FOR CONSTANT VELOCITY JOINTS



(57) Abstract: Urea grease composition for constant velocity joints, comprising base oil, one or more urea thickener compounds: A) one or more molybdenum sulphurized dialkyldithiocarbamates represented by general formula (I), wherein R^1 and R^2 are independently chosen from alkyl groups of from 1 to 24 carbons, m + n = 4, m is from 0 to 3 and n is from 4 to 1, B) triphenylphosphorothionate represented by formula (II) and C) one or more stearic acid metal salts.

UREA GREASE COMPOSITION FOR CONSTANT VELOCITY JOINTS

The present invention relates to a urea grease composition for constant velocity joints; more specifically, it relates to a urea grease composition for constant velocity joints which inhibits vibration and has a low friction coefficient.

The urea grease composition of the present invention is particularly suitable as a lubricant for plunging-type constant velocity joints in automobiles.

Constant velocity joints are universal joints which can transfer motive power from an automobile engine to the wheels while maintaining a constant angular velocity and torque. Whilst propeller shafts have been used in most automobiles, modern automobile design is following the trend for front-wheel drive, and there are many types of constant velocity joints permitting this front wheel drive. Constant velocity joints that are also plunging type joints are structured such that they slide in the axial direction; it is friction resistance to this sliding that causes the source vibration that results in vibration and noise within the automobile. Thus, there is a strong demand for a grease composition that is excellent in decreasing interior joint friction.

In order to treat this kind of problem, many examples of constant velocity joints have appeared on the market that use urea greases having excellent friction properties.

The technology disclosed in Japanese unexamined patent application H6-57283 is a grease composition for

constant velocity joints, where (a) molybdenum disulphide, (b) molybdenum sulphurized dialkyldithiocarbamate and (c) lead dialkyldithiocarbamate incorporated into urea grease.

The technology disclosed in Japanese examined patent publication H5-79280 is a grease for constant velocity joints obtained by using, in urea grease, both molybdenum dithiocarbamate and molybdenum dithiophosphate additives or by mixing zinc dithiophosphate into these organomolybdenum compounds.

The technology disclosed in Japanese unexamined patent application H6-330072 is a grease composition for constant velocity joints, where (A) molybdenum sulphurized dialkyldithiocarbamate and (B) triphenylphosphorothionate are both added to urea grease.

The technology disclosed in Japanese unexamined patent application H10-147791 relates to a grease composition for plunging type constant velocity joints, where molybdenum dithiocarbamate, sulphur-phosphorus-based extreme-pressure agent and oxidized wax calcium salt are mixed into urea grease.

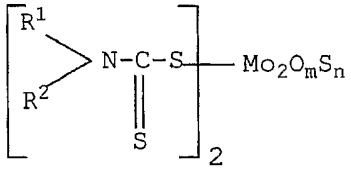
However, although the force generated in the axial direction of a plunging type constant velocity joint (hereinbelow, the driving force) is less than when commercial grease is used, these greases of the preceding art cannot be said to be low-friction force greases that are fully satisfactory for recent constant velocity joints used under severe conditions.

In the present invention, urea grease composition(s) have been developed for use in constant velocity joints, which grease compositions surprisingly exhibit advantageous decreases in the vibration generated in the

constant velocity joint, and improved low friction coefficients.

Accordingly, the present invention provides a urea grease composition for constant velocity joints, comprising base oil, one or more urea thickener compounds,

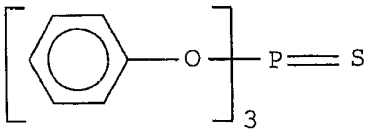
(A) one or more molybdenum sulphurized dialkyldithiocarbamates represented by general formula (I)



(I)

wherein R^1 and R^2 are independently chosen from alkyl groups of from 1 to 24 carbons, m + n = 4, m is from 0 to 3 and n is from 4 to 1,

(B) triphenylphosphorothionate represented by formula (II)



(II)

and

(C) one or more stearic acid metal salts.

In a preferred embodiment of the present invention, the urea grease composition further comprises (D) one or more calcium-based dispersants.

The urea grease composition of the present invention preferably comprises from 0.5 to 10% by weight of component (A) incorporated therein, with respect to the total weight of the urea grease composition.

The urea grease composition of the present invention preferably comprises from 0.1 to 10% by weight of component (B) incorporated therein, with respect to the total weight of the urea grease composition.

The urea grease composition of the present invention preferably comprises from 0.5 to 10% by weight of component (C) incorporated therein, with respect to the total weight of the urea grease composition.

The urea grease composition of the present invention preferably comprises from 0.5 to 10% by weight of component (D) incorporated therein, with respect to the total weight of the urea grease composition.

In a preferred embodiment of the invention, the one or more urea thickener compounds are present in the urea grease composition an amount in the range of from 2 to 35% by weight, with respect to the sum of the amounts of base oil and urea thickener compounds.

If there is less than 2% by weight of urea thickener compound(s), there may be little thickening effect and the system may not readily become greasy. If there is more than 35% by weight of urea thickener compound(s), the grease may become too hard and the lubricating effect may be unsatisfactory.

The urea grease composition used in the present invention is a grease composition comprising, as base oil, a lubricating oil such as mineral oil, ester-based synthetic oil, ether-based synthetic oil, hydrocarbon-based synthetic oil, or a mixture thereof, and, as thickener, a urea thickener compound obtained by reacting aliphatic amine, alicyclic amine, aromatic amine or the like with various isocyanate compounds, and there are no particular limitations.

Base oils of mineral origin may include those produced by solvent refining or hydroprocessing. Examples of mineral oils that may be conveniently used include those sold by member companies of the Royal Dutch/Shell Group under the designations "HVI", "MVIN", or "HMVIP".

Specific examples of synthetic oils include polyolefins such as α-olefin oligomer and polybutene, polyalkylene glycols such as polyethylene glycol and polypropylene glycol, diesters such as di-2-ethyl hexyl sebacate and di-2-ethyl hexyl adipate, polyol esters such as trimethylolpropane ester and pentaerythritol ester, perfluoroalkyl ethers, silicone oils, polyphenyl ethers, either individually or as mixed oils.

Base oils of the type manufactured by the hydroisomerisation of wax, such as those sold by member companies of the Royal Dutch/Shell Group under the designation "XHVI" (trade mark), may also be used.

Mineral oil and/or synthetic oil can be used as the base oil in the grease composition of the present invention.

Urea thickener compounds that may be conveniently used as thickeners include diurea, triurea, tetraurea and the like. Typical examples of diurea compounds can be obtained by reacting diisocyanate and monoamine; examples of diisocyanates include diphenylmethane diisocyanate, phenylene diisocyanate, diphenyl diisocyanate, phenyl diisocyanate and tolylene diisocyanate, and examples of monoamines include octylamine, dodecylamine, hexadecylamine, octadecylamine and oleylamine.

All these examples of thickeners and base oils can be used either individually or as mixtures, and there are

no particular limitations regarding these in the present invention.

Specific examples of the molybdenum sulphurized dialkyldithiocarbamate which may be conveniently employed as component (A) include molybdenum sulphurized diethyldithiocarbamate, molybdenum sulphurized dipropyldithiocarbamate, molybdenum sulphurized dibutyldithiocarbamate, molybdenum sulphurized dipentyldithiocarbamate, molybdenum sulphurized dihexyldithiocarbamate, molybdenum sulphurized didecyldithiocarbamate, molybdenum sulphurized diisobutyldithiocarbamate, molybdenum sulphurized di-(2-ethylhexyl)-dithiocarbamate, molybdenum sulphurized diamyldithiocarbamate, molybdenum sulphurized dilauryldithiocarbamate and molybdenum sulphurized distearyldithiocarbamate.

The amount of component (A) added is preferably in the range of from 0.5 to 10% by weight, more preferably in the range of from 0.5 to 5% by weight, with respect to the total weight of the urea grease composition.

If more than 10% by weight of component (A) is incorporated, the friction coefficient-lowering effect may not be as desired, and may become deleterious. If less than 0.5% by weight of component (A) is added, there may be no improvement in friction properties

The triphenylphosphorothionate that is employed as component (B) is preferably incorporated in the urea grease composition of the present invention in an amount in the range of from 0.1 to 10% by weight, more preferably in an amount in the range of from 0.1 to 5% by weight, with respect to the total weight of the urea grease composition.

If there is less than 0.1% by weight of component (B), there may be no improvement in friction or abrasion properties. However, if there is more than 10% by weight of component (B) it may not be possible to achieve adequate lubrication.

Examples of stearic acid metal salts that may be conveniently used in the urea grease composition of the present invention as component (C) include lithium stearate, calcium stearate, sodium stearate, barium stearate, magnesium stearate, aluminium stearate, zinc stearate.

Component (C) is preferably incorporated in the urea grease composition of the present invention in an amount in the range of from 0.5 to 10% by weight, more preferably in the range of from 0.5 to 5% by weight, with respect to the total amount.

If more than 10% by weight of component (C) is incorporated, there may be no lowering of the friction properties. Furthermore, the grease may become harder and it may be difficult to achieve the desired penetration. If less than 0.5% by weight of component (C) is incorporated, the friction property improvement may be unsatisfactory.

It should be noted that whilst there are many higher fatty acids other than stearic acid, and countless similar metal salts thereof, it has been surprisingly found in the present invention that only stearic acid metal salts can considerably lower the driving force of constant velocity joints. Other higher fatty acid metal salts do not result in adequate lowering of the driving force.

The calcium-based dispersants that may be conveniently used as component (D) are dispersants having calcium in the molecular structure, and dispersant refers to an additive that can disperse solids and the like in oil.

Typical examples thereof are calcium phenate, calcium sulphonate and calcium salicylate, although component (D) is not limited thereto.

Component (D) is preferably incorporated in an amount in the range of from 0.5 to 10% by weight, more preferably in the range of from 1 to 7% by weight, with respect to the total weight of the urea grease composition.

If less than 0.5% by weight of component (D) is added, there may be no adequate dispersion effect or lubrication, and if more than 10% by weight of component (D) is added, the grease may become soft.

In order to further improve the properties of the urea grease composition of the present invention, additional additives such as antioxidants, rust-preventing agents, extreme-pressure agents, polymers and the like can also be incorporated therein.

For example, antioxidants including alkylphenol, hindered phenol, alkylamine, diphenylamine and triazine antioxidants; anticorrosion agents including calcium sulphonate, sodium sulphonate, barium sulphonate and amino derivatives or metal salts of carboxylic acids; and extreme pressure agents including sulphurized oils or fats, sulphurized olefins, phosphoric acid esters, tricresyl phosphate, trialkyl thiophosphates and triphenyl phosphorothionates may be conveniently used.

Lubricants for constant velocity joints may

advantageously comprise the urea grease composition of the present invention.

Accordingly, the present invention further provides a method of lubricating a constant velocity joint comprising packing the constant velocity joint with the urea grease composition of the present invention; and a constant velocity joint packed with the urea grease composition of the present invention.

The present invention further provides the use of the urea grease composition of the present invention as a friction-reducing and vibration-reducing grease composition and, in particular, the use of said grease composition to reduce friction and/or vibration in a constant velocity joint.

The present invention is described below with reference to the following Examples, which are not intended to limit the scope of the present invention in any way.

EXAMPLES

The urea grease composition for constant velocity joints in the Working Examples and Comparative Examples below was obtained by adding additives to a urea base grease composition comprising base oil and urea thickener compound, in the proportions shown in Tables 1 to 5, and treating the resulting system using a three-roller mill. Purified mineral oil having a kinematic viscosity of 15 mm^2/s at 100°C was used as the afore-mentioned base oil.

The additives used in the Working Examples were zinc stearate and aluminium stearate, which are typical examples of component (C), and calcium salicylate and calcium sulphonate were used as component (D).

In Working Examples 1 to 5, 4 types of additive were incorporated into the urea base grease composition, i.e. component (A), component (B), component (C) and component (D).

In Working Example 6, component (A), component (B) and component (C) were incorporated into the urea base grease composition.

In Comparative Examples 1 to 6, two or three components selected from components (A), (B), (C) and (D) were incorporated into the urea base grease composition.

In Comparative Example 7, components (A) and (B) were incorporated into the urea base grease composition. The composition of said comparative Example corresponds to the composition disclosed in Japanese unexamined patent H6-330072.

The diurea grease in Tables 1 to 5 was obtained by reacting 2 mol of diphenylmethane diisocyanate with 2 mol of octylamine and 2 mol of oleyl amine, and uniformly dispersing the resulting urea thickener compound, in base oil. The resulting urea base grease composition had a penetration (25°C, 60 W) of 268, and a dropping point of 221°C. It should be noted that the urea thickener compound content therein was 10% by weight, with respect to the sum of the amounts of base oil and urea thickener compound.

The tetraurea grease in Tables 2 and 4 was obtained by reacting 2 mol of diphenylmethane diisocyanate with 1 mol of octylamine, 1 mol of laurylamine and 1 mol of ethylene diamine, and uniformly dispersing the resulting urea compound, in base oil. The resulting urea base grease composition had a penetration (25°C, 60 W) of 325, and a dropping point of 253°C. It should be noted that

the urea thickener compound content therein was 13% by weight, with respect to the sum of the amounts of base oil and urea thickener compound.

The penetration, dropping point and friction coefficient shown in the Tables were appraised by performing the following tests.

(1) Penetration

The penetration was measured according to the JIS K2220 penetration test method.

(2) Dropping point

The dropping point was measured according to the JIS K2220 dropping point test method.

(3) Friction coefficient

Of the numerous friction and abrasion test apparatuses, the SRV test apparatus, with a high correlation with driving force, was used; the friction coefficient was found under the following test conditions.

Test sample: bowl (Φ 17.5 mm)/plate
Load: 300 N
Slip speed: 0.18 m/s
Temperature: room temperature
Time: 10 min
Grease: approximately 1 g coating of grease on the test sample

Tables 1 to 5 also show an appraisal of whether the grease composition was satisfactory when used on a constant velocity joint, with the grease composition of Comparative Example 7 taken as the standard reference grease.

- ● Excellent driving force-reducing effect
- o Good driving force-reducing effect

□ No effect

× High driving force

*1 A is molybdenum sulphurized dialkyldithiocarbamate, the alkyl group is C4, n is a mixture of 2 and 3.

*2 B is triphenylphosphorothionate.

*3 C-1 is a stearic acid metal salt, the metal is zinc.

*4 C-2 is a stearic acid metal salt, the metal is aluminium.

*5 D-1 is a calcium-based dispersant, calcium salicylate.

*6 D-2 is a calcium-based dispersant, calcium sulphonate.

Table 1

Working Example		1	2	3
Base grease (% wt.)	Diurea grease	91.0	91.0	91.0
	Tetraurea grease	-	-	-
Additive (% wt.)	A *1	3.0	3.0	3.0
	B *2	2.0	2.0	2.0
	C-1 *3	2.0	2.0	-
	C-2 *4	-	-	2.0
	D-1 *5	2.0	-	2.0
	D-2 *6	-	2.0	
Penetration 60 W		275	276	272
Dropping point (°C)		216	214	212
Friction coefficient by SRV test		0.034	0.034	0.038
Satisfaction when used in constant velocity joint		●	●	○

Table 2

Working Example		4	5	6
Base grease (% wt.)	Diurea grease	91.0	-	93.0
	Tetraurea grease	-	91.0	-
Additive (% wt.)	A *1	3.0	3.0	3.0
	B *2	2.0	2.0	2.0
	C-1 *3	-	2.0	-
	C-2 *4	2.0	-	2.0
	D-1 *5	-	2.0	-
	D-2 *6	2.0	-	-
Penetration 60 W		272	336	338
Dropping point (°C)		215	242	241
Friction coefficient by SRV test		0.040	0.036	0.042
Satisfaction when used in constant velocity joint		o	•	o

Table 3

Comparative Example		1	2	3
Base grease (% wt.)	Diurea grease	93.0	95.0	95.0
	Tetraurea grease	-	-	-
Additive (% wt.)	A *1	3.0	-	-
	B *2	-	3.0	-
	C-1 *3	2.0	-	-
	C-2 *4			2.0
	D-1 *5	2.0	2.0	
	D-2 *6			3.0
Penetration 60 W		274	272	279
Dropping point (°C)		213	216	211
Friction coefficient by SRV test		0.061	0.120	Burnt
Satisfaction when used in constant velocity joint		x	x	x

Table 4

Comparative Example		4	5	6
Base grease (% wt.)	Diurea grease	93.0	-	93.0
	Tetraurea grease	-	95.0	-
Additive (% wt.)	A *1	3.0	3.0	3.0
	B *2	2.0	-	2.0
	C-1 *3	-	-	-
	C-2 *4	-	2.0	-
	D-1 *5	2.0	-	-
	D-2 *6	-	-	2.0
Penetration 60 W		272	328	298
Dropping point (°C)		217	246	211
Friction coefficient by SRV test		0.057	Burnt	0.070
Satisfaction when used in constant velocity joint		□	×	□

Table 5

Comparative Example		7
Base grease (% wt.)	Diurea grease	95.0
	Tetraurea grease	-
Additive (% wt.)	A *1	3.0
	B *2	2.0
	C-1 *3	-
	C-2 *4	-
	D-1 *5	-
	D-2 *6	-
Penetration 60 W		269
Dropping point (°C)		220
Friction coefficient by SRV test		0.071
Satisfaction when used in constant velocity joint		Standard

The grease compositions of Working Examples 1 to 5, which comprised component (A), component (B) component (C) and component (D) incorporated as additives, and the grease composition of Working Example 6, which was obtained using component (A), component (B) and component (C), afforded much lower friction coefficients than those

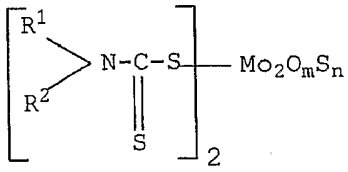
of Comparative Examples 1 to 7, which comprised any two or three of components (A), (B) and (C) as additives. Also, Working Examples 1 to 6 afforded a better decrease in driving force than Comparative Example 7, which was the known composition of Japanese unexamined patent H6-330072.

It is evident that the present invention provides a urea grease composition which can surprisingly inhibit vibration in constant velocity joints, and can considerably lower the friction coefficient.

C L A I M S

1. Urea grease composition for constant velocity joints, comprising base oil, one or more urea thickener compounds,

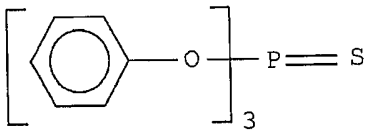
(A) one or more molybdenum sulphurized dialkyldithiocarbamates represented by general formula (I)



(I)

wherein R^1 and R^2 are independently chosen from alkyl groups of from 1 to 24 carbons, m + n = 4, m is from 0 to 3 and n is from 4 to 1,

(B) triphenylphosphorothionate represented by formula (II)



(II)

and

(C) one or more stearic acid metal salts.

2. Urea grease composition according to Claim 1, which grease composition further comprises (D) one or more calcium-based dispersants.

3. Urea grease composition according to Claim 1 or 2, wherein from 0.5 to 10% by weight of component (A) is incorporated therein, with respect to the total weight of the urea grease composition.

4. Urea grease composition according to any one of Claims 1 to 3, wherein from 0.1 to 10% by weight of

component (B) is incorporated therein, with respect to the total weight of the urea grease composition.

5. Urea grease composition according to any one of Claims 1 to 4, wherein from 0.5 to 10% by weight of component (C) is incorporated therein, with respect to the total weight of the urea grease composition.

6. Urea grease composition according to any one of Claims 1 to 5, wherein from 0.5 to 10% by weight of component (D) is incorporated therein with respect to the total weight of urea grease composition.

7. Urea grease composition according to any one of Claims 1 to 6, wherein from 2 to 35% by weight of the one or more urea thickener compounds are incorporated therein with respect to the sum of the amounts of base oil and urea thickener compounds.

8. A method of lubricating a constant velocity joint, comprising packing the constant velocity joint with the urea grease composition according to any one of Claims 1 to 7.

9. Use of the urea grease composition according to any one of Claims 1 to 7 to reduce friction and/or vibration in a constant velocity joint.

10. A constant velocity joint packed with the urea grease composition according to any one of Claims 1 to 7.

INTERNATIONAL SEARCH REPORT

International Application No
PCT/EP2004/051161

A. CLASSIFICATION OF SUBJECT MATTER
IPC 7 C10M169/06

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)
IPC 7 C10M

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.
Y	EP 0 633 304 A (TOYOTA MOTOR CO LTD ; SHOWA SHELL SEKIYU (JP)) 11 January 1995 (1995-01-11) cited in the application	1,3-5, 7-9
X	page 6, line 16 - line 25; claim 1	10
Y	US 6 037 314 A (ISHII HITOSHI ET AL) 14 March 2000 (2000-03-14) column 1, lines 45-49	1-9
X	column 3, line 56 - column 4, line 26; claim 1	10
A	WO 01/64821 A (NIMURA TETSUO ; SAITO KOUJI (JP); WADA TOMOYASU (JP); SHINODA NORIAKI) 7 September 2001 (2001-09-07)	1,3-5, 7-9
X	claims 1,6,10	10

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[X] Further documents are listed in the continuation of box C.

[X] Patent family members are listed in annex.

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Date of the actual completion of the international search	Date of mailing of the international search report
18 October 2004	29/10/2004

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INTERNATIONAL SEARCH REPORT

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C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category °	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	EP 0 714 975 A (SHOWA SHELL SEKIYU) 5 June 1996 (1996-06-05)	1,3-5, 7-9
X	page 1, line 5 - line 7; claim 1	10
Y	EP 0 761 806 A (LUBRIZOL CORP) 12 March 1997 (1997-03-12) page 9, line 16 - line 58; claim 1	1-9

INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No
PCT/EP2004/051161

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
EP 0633304 A	11-01-1995	JP 3001171 B2	24-01-2000
		JP 6330072 A	29-11-1994
		BR 9402060 A	27-12-1994
		CA 2124283 A1	26-11-1994
		CN 1097796 A ,B	25-01-1995
		DE 69408604 D1	02-04-1998
		DE 69408604 T2	16-07-1998
		EP 0633304 A1	11-01-1995
		ES 2114086 T3	16-05-1998
		HK 1009341 A1	28-05-1999
		KR 9707496 B1	09-05-1997
		US 5449471 A	12-09-1995
US 6037314 A	14-03-2000	JP 9324190 A	16-12-1997
		DE 19723960 A1	11-12-1997
WO 0164821 A	07-09-2001	JP 2001240886 A	04-09-2001
		WO 0164821 A2	07-09-2001
		AU 4417801 A	12-09-2001
		BR 0108775 A	26-11-2002
		CA 2401507 A1	07-09-2001
		EP 1265977 A2	18-12-2002
		US 2003027728 A1	06-02-2003
		CN 1406270 T	26-03-2003
		ZA 200206944 A	30-04-2003
EP 0714975 A	05-06-1996	JP 8157859 A	18-06-1996
		AU 696648 B2	17-09-1998
		AU 3919595 A	13-06-1996
		BR 9505614 A	16-09-1997
		CA 2164053 A1	03-06-1996
		CN 1132781 A ,B	09-10-1996
		DE 69502164 D1	28-05-1998
		DE 69502164 T2	24-09-1998
		EP 0714975 A1	05-06-1996
		ES 2116030 T3	01-07-1998
		HK 1010459 A1	17-06-1999
		US 5624889 A	29-04-1997
EP 0761806 A	12-03-1997	AU 721723 B2	13-07-2000
		AU 6194496 A	27-02-1997
		CA 2183811 A1	25-02-1997
		DE 69616458 D1	06-12-2001
		DE 69616458 T2	11-07-2002
		EP 0761806 A1	12-03-1997
		ES 2166867 T3	01-05-2002
		JP 9104889 A	22-04-1997
		SG 72700 A1	23-05-2000
		US 6214778 B1	10-04-2001